

THESIS FOR THE DEGREE OF LICENTIATE OF ENGINEERING

Gentle Remediation Options (GRO) for Managing Risks and Providing
Ecosystem Services at Contaminated Sites

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Cover:

Generic GRO risk management framework diagram, developed in Drenning et al. (2022).

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ABSTRACT

Soils are a non-renewable resource and comprise a key component of the world's stock of natural capital. Due to industrialisation, urbanisation and other patterns of unsustainable development, widespread land degradation in the form of contamination, soil sealing, compaction, etc. has impaired the capacity of soils to perform their essential functions and provide humans with vital ecosystem services. Brownfields are typically urban or peri-urban sites that have been affected by the former uses of the site, are or are perceived to be contaminated, and require intervention to bring them back to beneficial use. They also constitute an important and underutilised land and soil resource to provide ecosystem services in urban areas as an element of green infrastructure through the use of nature-based solutions such as gentle remediation options (GRO). Gentle Remediation Options (GRO) are remediation measures involving plants, fungi, bacteria, and soil amendments that can be applied to manage risks at contaminated sites. Several studies and decision-support tools promote the wider range of benefits provided by GRO, including improving soil function to provide ecosystem services, but there is still scepticism regarding GRO implementation. Interviews with a small group of experts have elucidated some of the main possibilities and challenges for GRO implementation in Sweden. As a result, a risk management framework for GRO has been developed to strengthen the decision basis for GRO implementation in practice and address some of the key issues that need to be better communicated, including the various risk mitigation mechanisms, the required risk reduction for an envisioned land use, and the time perspective associated with the risk mitigation mechanisms. The framework is envisioned to be used as a tool for risk communication with stakeholders, decision-makers and regulatory agencies to identify GRO strategies for managing risks at contaminated sites and supporting phytomanagement for sustainable remediation and development. Two case studies are used to demonstrate the application of the risk management framework: Polstjärnegatan and Kolleberga.

Keywords: Brownfields; Gentle Remediation Options (GRO); Phytomanagement; Ecosystem Services; Risk-Based Land Management (RBLM); Sustainable Remediation

LIST OF PUBLICATIONS

This thesis contains the following publications appended to the thesis:

- I. Drenning, P., Norrman, J., Chowdhury, S., Rosén, L., Volchko, Y., Andersson-Sköld, Y. (2020). Enhancing ecosystem services at urban brownfield sites - What value does contaminated soil have in the built environment? IOP Conf. Ser. Earth Environ. Sci. 588. <https://doi.org/10.1088/1755-1315/588/5/052008>
- II. Drenning, P., Chowdhury, S., Volchko, Y., Rosén, L., Andersson-Sköld, Y., Norrman, J. (2022). A risk management framework for Gentle Remediation Options (GRO). Sci. Total Environ. 802. <https://doi.org/10.1016/j.scitotenv.2021.149880>

Division of work between the authors

Paper I: The authors contributed as follows: Conceptualization: P.D., S.C., Y.V., J.N.; Methodology: P.D., S.C., Y.V., J.N.; Investigation: P.D.; Visualization: P.D., S.C., Y.V., J.N.; Writing—original draft preparation: P.D., S.C., Y.V., J.N.; Writing—review and editing: P.D., S.C., Y.V., L.R., Y.A-S., J.N.; Supervision: Y.V., L.R., Y.A-S., J.N.; Project administration: J.N.; Funding acquisition, Y.V., L.R., J.N. All authors have read and agreed to the published version of the manuscript.

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- Drenning (2021a) *Gentle Remediation Options (GRO): A literature review (Part 1/2)*. Gothenburg, Sweden.
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- Drenning (2021b) *Soil Functions and Ecosystem Services: A literature review (Part 2/2)*. Gothenburg, Sweden.
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The bulk of the work towards the licentiate thesis took place during the height of the covid-19 pandemic and I would like to say a big thank you to the family, friends and co-workers who kept me sane throughout this difficult period. Existential stress is not the best companion for productive deep work, but I was able to manage thanks to the support I received from many caring people. Not least of which are my family and friends (Mellons) in the USA who are always caring and supportive (even if they have no idea why I'm still on the wrong side of the Atlantic Ocean), and to friends here in Gothenburg who make this place feel like home. And, of course, I am thankful every day for the love and support from my sambo, Annija (probably the main reason I am still sane), as well as the happy chance that brought us together at the very start of this otherwise dreadful pandemic.

Gothenburg, October 2021

Paul Drenning

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1 INTRODUCTION

This chapter provides a brief background of the research, presents the research aim and the main objectives of this thesis, as well as the scope of work, followed by clarifying limitations.

1.1 Background

A series of agenda-setting reports by the European commission (e.g. *Vision for a Resource Efficient Europe*, *European Biodiversity Strategy to 2020 & 2030*, *Zero Pollution Action Plan*, *European Green New Deal*) have raised awareness of the widespread degradation of ecosystems by over-exploitation, land-use change, contamination, sealing, compaction, erosion, neglect, etc. which have led to rapid losses in biodiversity and diminished the total provided ecosystem services by approximately 60% worldwide in the past 50 years alone (EC, 2019, 2011a, 2011b, 2006; Ellen MacArthur Foundation, 2015). Soil and its functions (Figure 1-1) have been raised to a position of critical importance for our common future through the *Thematic Strategy on Soil Protection* (EC, 2006), and is currently being updated and expanded within the scope of the *European Green New Deal* and *Biodiversity Strategy to 2030*. Within the *Thematic Strategy*, seven essential SF have been established: (i) biomass production, including agriculture and forestry; (ii) storing, filtering and transforming nutrients, substances and water; (iii) biodiversity pool, such as habitats, species and genes: (iv) physical and cultural environment for humans and human activities; (v) source of raw materials; (vi) acting as a carbon pool; (vii) archive of geological and archaeological heritage (EC, 2006). The significance of soil functions (SF) and soil-based ecosystem services (ES) for realising the UN's Sustainable Development Goals (SDG) has also been addressed by directly linking them to many of the SDGs (e.g. S. Keesstra et al. 2018; S. D. Keesstra et al. 2016).

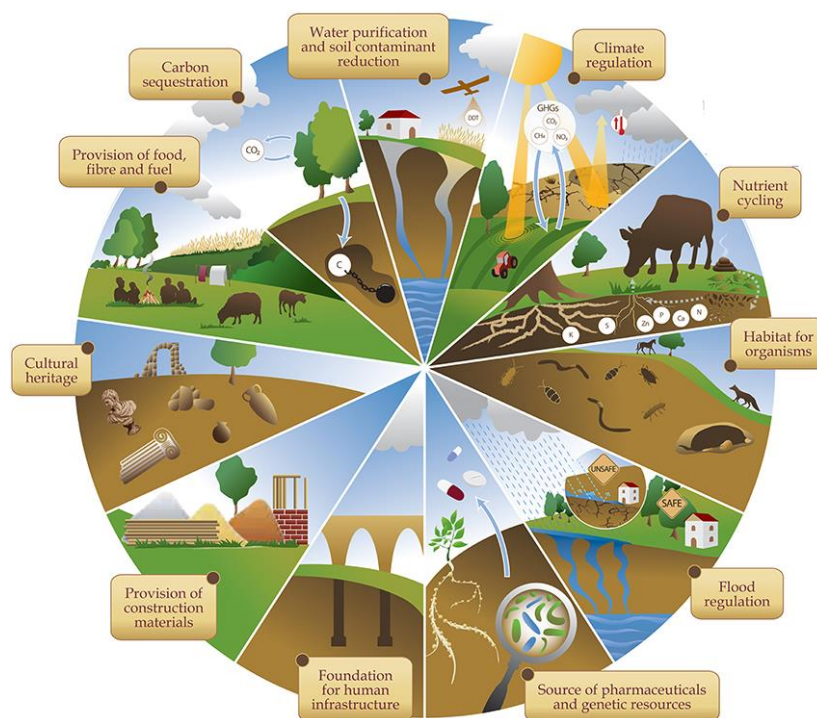


Figure 1-1. Schematic diagram of soil functions from the FAO, from Baveye, Baveye, and Gowdy 2016 (CC-BY 4.0).

Brownfields are underused or derelict areas with, in many cases, real or perceived, soil and groundwater contamination that require intervention to bring them back to beneficial use, which is often a barrier to redevelopment in terms of investment risks, ownership constraints, risk of future liability claims and public stigma (Ferber et al., 2006; ISO, 2017; Norrman et al., 2016; Vegter et al., 2002). In Europe, there are more than 2.5 million potentially contaminated sites caused by anthropogenic activity, i.e. brownfields, of which approximately 85,000 are in Sweden (Panagos et al., 2013; SEPA, 2021). Conventional soil remediation techniques are those that utilise physical, chemical, biological or a combination of methods to, most often, address the source of contamination (Kuppusamy et al., 2016b, 2016a; Swartjes, 2011). Remediation, however, is not intrinsically sustainable (Bardos et al., 2020a; Cundy et al., 2016). A common issue with many remediation techniques, especially ex-situ measures involving excavation but also some in-situ techniques, is that they can have considerable negative impacts. They may result in significant degradation or even elimination of the soil ecosystem and its essential functions, thereby rendering a soil unsuitable for 'soft' end uses like green spaces which require ecological functioning (Bardos et al., 2016; FAO et al., 2020; Gerhardt et al., 2017; Swartjes, 2011; Volchko et al., 2014b). In Sweden, remediation by soil excavation and landfilling ('dig-and-dump') or ex-situ treatment is the most commonly used method in practice since it is fast and effective for source removal, thus gaining regulatory approval, but is often the result of oversimplified, generic risk assessments coupled with conservatively applied legislative guidelines (SEPA, 2018a; SGI, 2018). There is, however, a recognized need for innovation and development, with fully 78% of practitioners in Sweden indicating a large need, of alternative remediation methods to prevent such 'over-remediation' and overuse of dig-and-dump (SEPA, 2018a; SGI, 2018). New practices are indeed crucial for sustainable remediation and brownfield regeneration, because a significant amount of brownfield land area remains derelict or underutilized due to rehabilitation being uneconomic or unsustainable using conventional methods (Bardos, 2014; Bardos et al., 2016). While it may be suitable for highly contaminated sites and hotspots, excavation is highly energy-intensive, costly and practitioners frequently fail to consider the irreversible damage removing soil layers can do to the environment. The necessity of such remediation is questionable for many applications, particularly if remediation is triggered due to unacceptable ecological risks and may be more damaging to the soil ecosystem than the contaminants themselves (FAO et al., 2020; Swartjes, 2011). Regrettably, contaminated soil has long been viewed as waste to be disposed of rather than as a valuable resource to be treated and reused (Gerhardt et al., 2017; Mench et al., 2010). "Green" alternatives to conventional soil remediation are gentle remediation options (GRO), which are in-situ remediation measures that utilise plants, fungi, bacteria, and soil amendments to break contaminant linkages. GRO may be viable alternatives to conventional techniques, in particular for large areas and contaminated sites that pose low to medium risks to human health and the environment (Andersson-Sköld et al., 2014; Cundy et al., 2016; Enell et al., 2016; GREENLAND, 2014a). Increasingly, research and successful application is showing that GRO can provide both effective risk management and a net gain in ecological soil function; nevertheless, widespread adoption is still lacking due to perceived (and actual) limitations, uncertainties and challenges (Cundy et al., 2016; Gerhardt et al., 2017; Mench et al., 2010; Vangronsveld et al., 2009).

1.2 Aim and objectives

The overall aim of this thesis is:

to support decision-makers by clarifying the challenges and possibilities for implementing GRO for risk management and as viable strategies for sustainable remediation and development of brownfields.

To reach the overall aim, the thesis has the following specific objectives:

- i. to highlight and discuss the benefits offered by GRO and the current frameworks of sustainable remediation to fit into the context of sustainable development;
- ii. to gain a better understanding of the challenges inherent to implementing GRO in Sweden;
- iii. to develop a framework for identifying GRO and communicating their potential for managing contamination risks to both human health and the environment; and
- iv. to demonstrate the developed framework in two case studies.

1.3 Scope of work

To achieve the aim and fulfil the specific objectives of this licentiate thesis, a multi-disciplinary approach was required. Identifying and exploring the intersection between related (yet often disconnected) fields – including contamination and associated risks, remediation of contaminated sites (both gentle and conventional), sustainability in remediation, soil science (requiring a study of soil biota, soil functioning and soil quality assessment) and associated fields – forms the groundwork for this Ph.D.-work. To establish the context within which this work has been carried out, the thesis begins with a theoretical background (Chapter 2) to briefly present the relevant topics.

The methodology section (Chapter 3) describes the process to achieve the aim of this licentiate thesis and lists the main steps followed to fulfil the research objectives. Results (Chapter 4) are provided in the subsequent section and include the following:

- Section 4.1 – Results on possibilities and benefits of GRO for sustainable remediation and development.
- Section 4.2 – Results from interviews with various experts in the field of remediation in Sweden. Anonymised answers to interviews with experts are summarised in a table format with additional points of interest from the discussions highlighted in the respective section.
- Section 4.3 – A risk management framework developed for gentle remediation options (GRO). The framework is presented as an illustration and further information is provided regarding i) a conceptualisation of linkages between land use, soil contaminants and time expectations when applying GRO and ii) the identification of GRO risk mitigation mechanisms via literature review.
- Section 4.4 – Application of the developed framework for two case study sites – Polstjärnegatan and Kolleberga in Sweden.

Following the results, a discussion (Chapter 5) of the licentiate thesis is provided in the next section to discuss broader implications including how GRO are viable strategies for sustainable remediation and development. Then, main conclusions (Chapter 6) that can be drawn from this

licentiate thesis are briefly summarised. In the final section, an outline of on ongoing and future work (Chapter 0) is given.

1.4 Limitations

The limitations of the licentiate thesis are as follows:

- As it is a multidisciplinary research, the focus has been put in linking different fields of interest rather than an in-depth exploration of each topic. Thus, the thesis provides a necessarily limited investigation into each of these, but the possible extent of the research is outlined as ongoing and future work in Chapter 7.
- GRO, phytomanagement, ecosystem services, soil science and brownfield redevelopment and regeneration are concepts with a solid scientific foundation but are developing quickly. Some important information may have been missed in writing this thesis and new material is being published regularly that may be concurrent with the writing of this thesis and not included here.
- Globalizing the essential knowledge pertaining to GRO in terms of risk management to create the proposed generic risk management framework inevitably led to some oversimplifications and are noted in the Discussion and Conclusions section. It is acknowledged that actual field application of GRO is a site-specific process that requires a more detailed risk assessment and in-depth knowledge of the site conditions to effectively manage the exposure risks to receptors at a contaminated site using GRO.

2 THEORETICAL BACKGROUND

This chapter briefly presents different concepts related to the research, connects them to support the proposal herein, and builds on the findings to elaborate upon the scope of the research.

2.1 Soil biology, functioning and ecosystem services

Soils make up a crucial part of the Earth's system and play fundamental roles in its functioning, upon which humans are dependent, as well as linking the atmosphere, the subsurface, and the aquatic realms (Barrios, 2007; Faber et al., 2013; Kibblewhite et al., 2008; Ritz et al., 2009). This section will discuss some of the essential aspects of biodiversity driving the ecological functioning of soils, how they can be grouped into understandable and measurable entities, which ecosystem services can be attributed to them and how they can be assessed. The field of soil biology, function and ecosystem services is vast and many concepts will be covered here in limited depth; for more information the reader is referred to Drenning (2021b) and other more extensive, in-depth reports e.g. (FAO et al., 2020; Orgiazzi et al., 2016; Turbé et al., 2010).

2.1.1 Soil biota

When referring to the soil system, it is common to emphasize the physical or material geochemistry (i.e. abiotic) component and neglect the living organisms (i.e. biotic) that are ultimately responsible for the majority of soil processes (Creamer et al., 2016; Doran and Zeiss, 2000; Griffiths et al., 2016; Kibblewhite et al., 2008; Ritz et al., 2009). Ritz et al. (2009) state that the physical (e.g. texture, bulk density, porosity, and water availability) and chemical (e.g. pH, organic matter content, metal availability) properties of soils provide the fundamental context, and set the limits, in which the biotic assemblages operate. Hence, they have a clear utility in assessing ecological status; however, the majority of soil processes are in fact driven by the soil biota. According to Kibblewhite et al. (2008), the unique and crucial feature of the soil organisms is that they are *adaptive to changes in environmental circumstances, driven by processes of natural selection*, in ways that the abiotic systems of the soil are not. Soil biota can be broadly separated by size into the following groupings: microbes/microflora, microfauna, mesofauna, macrofauna and megafauna. The organisms that can be included in these broad groupings and their associated roles in the soil system are briefly presented in Table 2-1.

Table 2-1. Soil biota organised by size class, summarised from (Wurst et al., 2013). Predominant organisms per group are in bold.

Size class	Dominating organisms	Soil processes	Associated functions and services
Microbes/ microflora	Bacteria, fungi , archaea	Degradation of organic matter, nitrogen fixation and denitrification, soil aggregation	Decomposition, carbon and nutrient cycling, disease suppression, regulation of plant growth and primary productivity
Microfauna	Nematodes , protozoa	Predation, herbivory, bacteriovory, fungivory parasitism, provide food source to other organisms, distribute microbes in rhizosphere	Nutrient cycling, regulation of population sizes, pest and disease suppression
Mesofauna	Mites, collembola (springtails), enchytraeids (potworms)	Herbivory, bacteriovory, fungivory, predation, provide food source to other organisms, distribute microbes in rhizosphere	Nutrient cycling, regulation of population sizes, pest and disease suppression
Macrofauna/ megafauna	Earthworms , ants, termites, spiders, millipedes, beetles, moles	Degradation of organic matter, predation, herbivory, parasitism, burrowing, soil mixing, soil aggregation, provide food source to other organisms	Decomposition, carbon and nutrient cycling, water regulation, pest and disease suppression, regulation of population sizes, positive/negative effects on plant growth and primary productivity

2.1.2 Soil functions and ecosystem services linkages

'Soil functions' is a loaded term which has been used alternatively to mean process, function, role, or service (Baveye et al., 2016; Bünemann et al., 2018), and is often used interchangeably with '(soil-based) ecosystem services' and 'ecosystem functions'. Confusing as the term may be, it has served as a conceptual foundation in soil management, most notably in the *Thematic Strategy* (EC, 2006), so it is considered worthwhile to clarify the terminology (Baveye et al., 2016). Accordingly, 'soil functions' are here defined as *what the soil has the capability to do in its natural (undisturbed) state as a result of the (bundles of) soil processes (e.g. soil formation, nutrient cycling, etc.) arising out of the complex interaction between biotic and abiotic components in the soil environment* (Bünemann et al., 2018; Orgiazzi et al., 2016; Volchko et al., 2013). In simple terms: 'soil functions' is used to define the biological, geochemical and physical processes and components that take place within a soil or larger ecosystem (i.e. underlying processes maintaining the ecosystem) and 'ecosystem services' encompasses the tangible and intangible benefits that humans obtain from ecosystems (Bünemann et al., 2018; Orgiazzi et al., 2016; Volchko et al., 2013).

Soil functionality and the delivery of ecosystem services is dependent on a healthy, living soil ecosystem; 'soil health' here referring to *the capacity of soil to function as a vital living system, within ecosystem and land-use boundaries, to sustain plant and animal productivity, maintain or enhance water and air quality, and promote plant and animal health* (Doran and Zeiss, 2000). It is generally understood that ecosystem services for human benefit are ultimately functional outputs of biological processes resulting from highly complex interactions between the soil biota and the abiotic physical and chemical environment of the soil (Kibblewhite et al., 2008). In aggregate, these soil (or ecosystem) functions are provided by assemblages of interacting organisms (i.e. specific groups of the soil biota) (Brussaard, 2013; Kibblewhite et

al., 2008), see Figure 2-1. Due to their perceived associations with certain ecosystem functions, groups of related biota interacting with each other and carrying out biological processes that contribute to these aggregate functions are often combined into so-called 'functional groups' or 'functional assemblages' (Brussaard, 2013; Kibblewhite et al., 2008). An important note is that these assemblages do not operate in isolation, but are part of an interactive and interdependent soil system (Kibblewhite et al., 2008; Pulleman et al., 2012; Wurst et al., 2013), and these relatively broad classifications are themselves generalisations since multiple functions can be performed by different functional assemblages and overlaps in biological processes occur across all levels (Pulleman et al., 2012), a concept which is broadly referred to as 'ecological multifunctionality' (Birgé et al., 2016; FAO et al., 2020; Wall et al., 2004).

Kibblewhite et al. (2008) synthesised the complex relationships between organisms by establishing four functional assemblages made up of 'key functional groups': 1) decomposers, 2) nutrient transformers, 3) ecosystem engineers, and 4) biocontrollers. Via their associated biological processes and functional attributes, the functional assemblages directly contribute to four key aggregate ecosystem functions that can in turn be linked directly to ecosystem services (Figure 2-1). The authors propose that overall soil health is "a direct expression of the condition of these assemblages, which in turn, depends on the physical and chemical condition of the soil habitat" (Kibblewhite et al., 2008).

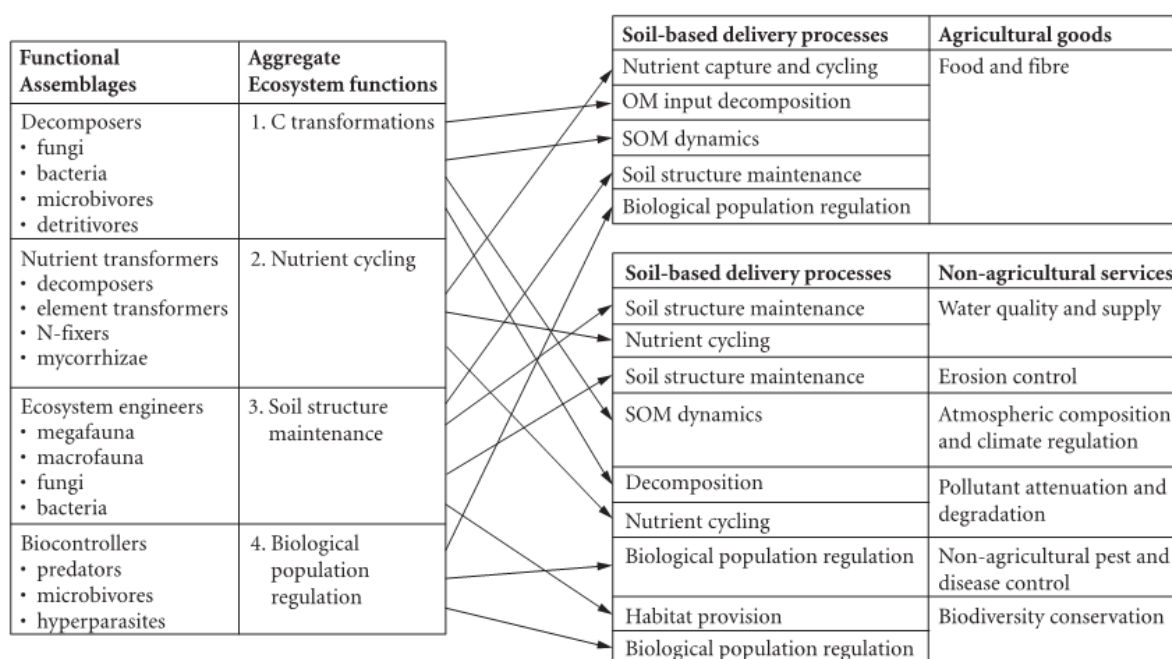


Figure 2-1. Conceptual framework of linkages between soil biota, biologically-mediated soil processes and the provision of soil-based ecosystem goods and services, from (Barrios et al., 2012) (adapted from (Kibblewhite et al., 2008))

As shown in Figure 2-1, the authors reason that soil health is fully dependent upon the maintenance of four key functions (i.e. bundles of processes aggregated into ecosystem functions): 1) Carbon (C) transformations – transformation of carbon through the

decomposition of plant residues and other organic matter together with the synthetic activities of the soil biota, including, and particularly, soil organic matter (SOM) synthesis; 2) Nutrient cycling – including nitrogen, phosphorous and sulphur and regulation of nitrous oxide emissions; 3) Soil structure maintenance – maintenance of the structure and fabric of the soil by aggregation and particle transport, and formation of biostructures and pore networks across many spatial scales by the combined action of plant roots and soil organisms commonly known as 'soil ecosystem engineers'; and 4) Biological population regulation – biological regulation of soil populations by competition, predation and parasitism, including organisms recognized as pests and diseases of agriculturally important plants and animals as well as humans (Kibblewhite et al., 2008). These aggregated ecosystem functions participate in more than one soil-based delivery process, and one or more soil-based delivery processes are required in-turn for the provision of ecosystem goods and services in agricultural landscapes (Barrios et al., 2012).

Classifications of soil organisms can be based on different criteria, and various levels of aggregation have been used between functional approaches (e.g. (Barrios, 2007; Kibblewhite et al., 2008; Wurst et al., 2013)). Addressing this issue, Turbé et al.(2010) divided the soil organisms according to three 'all-encompassing ecosystem functions': 1) transformation and decomposition (i.e. a combination of carbon transformations and nutrient cycling), 2) biological regulation and 3) soil engineering (i.e. soil structure maintenance) (Turbé et al., 2010). Each of these functions can be performed by assemblages of soil organisms separated into just three broad functional groups (overlapping with those mentioned previously in (Kibblewhite et al., 2008; Wurst et al., 2013)):

Ecosystem engineers – earthworms, enchytraeids, ants, termites and some small mammals modify or create habitats for smaller soil organisms by building resistant soil aggregates and pores. In this way, they also regulate the availability of resources for other soil organisms since soil structures become hotspots of microbial activities.

Chemical engineers – includes microorganisms (the most abundant soil species) such as bacteria, fungi and protozoans that are responsible for carbon transformation through the decomposition of plant residues and other organic matter as well as transformation of nutrients (e.g. nitrogen, phosphorous, sulphur) made readily available for plants, animals and humans.

Note: this group is a combination of *decomposers* and *nutrient transformers*.

Biological regulators – comprises a large variety of small invertebrates, such as nematodes, pot worms, springtails, and mites, which act as predators of plants, other invertebrates, or microorganisms by regulating their dynamics in space and time.

Although not technically part of the soil biota, vegetation also plays a key role in the soil ecosystem and performs certain biological processes that can greatly influence soil organisms. Brussaard (2013) highlights two biological processes of particular importance in soils: *photosynthesis* (i.e. composition/C fixation, largely occurring aboveground, associated with plant growth) and *respiration* (i.e. decomposition/ C dissipation, largely occurring belowground, inasmuch as associated with plant death). Recognising carbon (C) as the common

denominator ('common currency') and main factor that integrates ecosystem functions implies that the concept of soil functional groups being responsible for ecosystem processes that result in ecosystem services cannot be discussed without accounting for a link to the vegetation (Brussaard, 2013). This view stresses the importance of *primary productivity* (the rate of energy capture and carbon fixation by primary producers) as a driver of ecosystem processes and a key determinant of soil biodiversity (Brussaard, 2013; Prosser et al., 2007; Turbé et al., 2010; Wall et al., 2012). Furthermore, both the abundance and the health of vegetation are intricately linked to the diversity of functions performed by soil biota, since the functional groups contribute to the availability of nutrients and to the soil structure, two crucial parameters for plant growth (Turbé et al., 2010). The inverse is also true (i.e. interdependence), and there have been demonstrable positive effects by vegetation on the soil as a habitat for organisms even creating 'hotspots' of biological activity due to greater availability of C substrates (Barrios, 2007).

2.1.3 Soil-based ecosystem services

Many ecosystem services can be intuitively linked to the functioning of the soil biota and their interactions within their physical and chemical environment (Brussaard, 2013; Dominati et al., 2010; Faber and Van Wensem, 2012; Orgiazzi et al., 2016; Thomsen et al., 2012). Extensive lists of soil-based ecosystem services have been covered by many different authors (e.g. (Brussaard, 2013; Dominati et al., 2010; FAO et al., 2020; Haygarth and Ritz, 2009; Orgiazzi et al., 2016; Robinson et al., 2013; Wall et al., 2004)) with both considerable differences in terminology and overlap between the many variations.

Most of the ecosystem services provided by soils are supporting services, or services that are not directly used by humans, but underlie the provisioning of all other services (Turbé et al., 2010). These include nutrient cycling, soil formation and primary production. In addition, soil biodiversity influences the main regulatory services, namely the regulation of atmospheric composition and climate, water quantity and quality, pest and disease incidence in agricultural and natural ecosystems, and human diseases. Soil organisms may also control or reduce environmental pollution (e.g. via bioremediation). Soil organisms also contribute to provisioning services that directly benefit humans, for example, genetic resources of soil microorganisms used for developing pharmaceuticals. According to Turbé et al. (2010), the contributions of soil biodiversity, in terms of soil-based ecosystem services, can be grouped under the six following aggregated categories: 1) Soil structure, soil organic matter and fertility, 2) Regulation of carbon flux and climate control, 3) Regulation of the water cycle, 4) Decontamination and bioremediation, 5) Pest control, and 6) Human health.

Bünemann et al. (2018) recommend that to better consolidate the ecosystem services throughout these various schemes and frameworks, they can be seen as a soil-related sub-set of the ecosystem services according to the Common International Classification of Ecosystem Services (CICES¹) or a similar classification system (Bünemann et al., 2018). For example, one of the clearest delineations of soil-based ecosystem services (based on the Millennium

¹ <https://biodiversity.europa.eu/maes/common-international-classification-of-ecosystem-services-cices-classification-version-4.3>

Ecosystem Assessment) is presented in the Global Soil Biodiversity Atlas (Orgiazzi et al., 2016) by linking them to correlated soil/ecosystem functions and soil biota (Figure 2-2), which was derived from the conceptual framework shown in Figure 2-1.

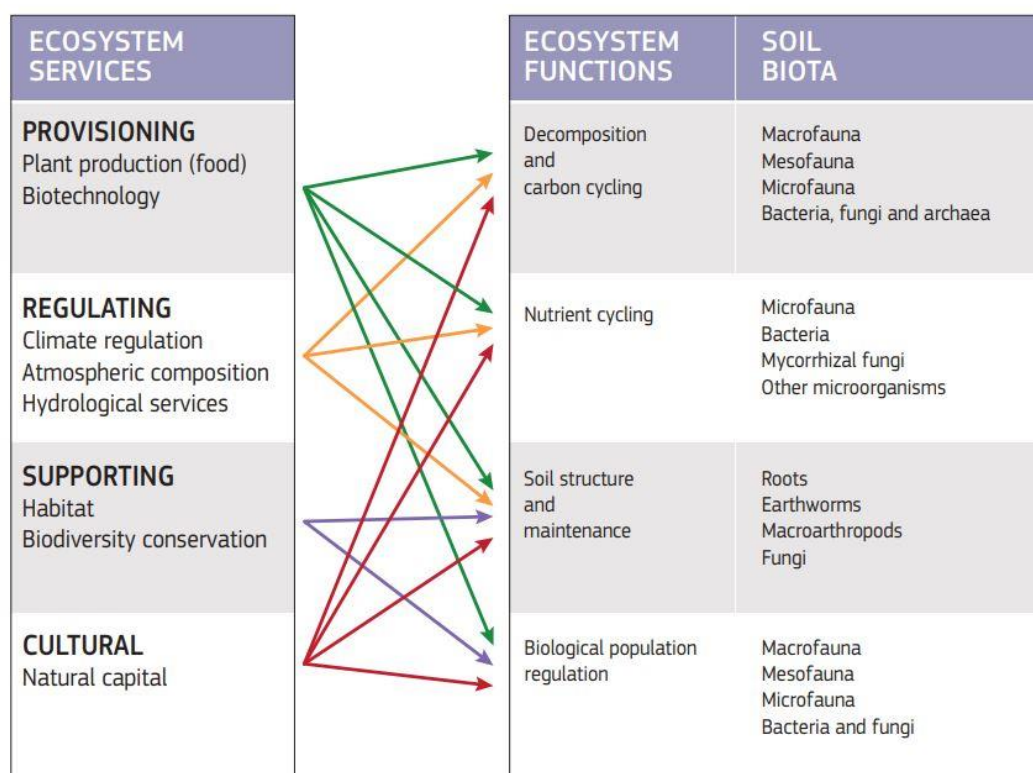


Figure 2-2. Soil-based ecosystem services, ecosystem functions and soil organisms that support them, from (Orgiazzi et al., 2016).

2.1.4 Soil quality and ecosystem service assessment

'Soil quality' refers generally to *the capacity of a soil to perform its functions as necessary for its intended end use* (Bünemann et al., 2018; Garbisu et al., 2011; Karlen et al., 2003, 1997; USDA Natural Resource Conservation Service, 2015; Volchko et al., 2013). This inherently anthropocentric definition has also been expanded to more broadly include ecological (i.e. biological) functioning *within ecosystem and land-use boundaries to sustain biological productivity, maintain environmental quality, and promote plant and animal health* (Bünemann et al., 2018; Orgiazzi et al., 2016). This expanded definition encompasses 'soil health' and better reflects the complexity and site-specificity of soil functioning as well as indicating the multi-functionality of soils when functioning according to their capacity. A key aspect of soil quality is that it is assessable through the use of soil quality indicators (SQI), which are *measurable properties of the soil used to evaluate the degree to which the soil quality matches the soil functions determined by the intended end use of the soil* (Bünemann et al., 2018; Volchko et al., 2013).

There are many published methodologies for soil quality (or health, function or services) assessment for various purposes (see Pulleman et al., (2012) for an overview of European

approaches) using a wide range of SQI, e.g. (Andrews et al., 2004; Epelde et al., 2014b; Gugino et al., 2009; Moebius-Clune et al., 2016; Rutgers et al., 2012; Thomsen et al., 2012; Velasquez et al., 2007; Volchko et al., 2019a, 2014b). The methodology and indicators used vary but the common aim is to identify and measure biotic or abiotic characteristics that are correlated (or at least thought to be) with soil functions and ecosystem services of interest (Baveye et al., 2016; Bünemann et al., 2018). Multiple criteria addressing these issues (e.g. meaningfulness, standardisation, measurability and cost-efficiency, sensitivity/accuracy, etc.) have commonly been applied to filter the extensive range of potential SQI to select those most suitable (e.g. (Bünemann et al., 2018; Doran and Zeiss, 2000; Faber et al., 2013; Griffiths et al., 2016; Gutiérrez et al., 2015; Ritz et al., 2009; Stone et al., 2016; Turbé et al., 2010)). No single indicator will comply with all these criteria, so, in practice, efforts have been placed on the development of sets of complementary indicators, including both biotic and abiotic parameters, as selected by users (Pulleman et al., 2012; Turbé et al., 2010).

Assessment and monitoring of soil quality has focused mainly on abiotic, physico-chemical soil properties as indicators (e.g. pH, organic matter content, CEC, nutrient availability, water capacity, soil texture, etc.), but biological parameters are becoming increasingly used in soil quality assessments as they can provide a more direct measure of soil functioning (Alkorta et al., 2003; Bünemann et al., 2018; Epelde et al., 2009a; Faber et al., 2013; Garbisu et al., 2011; Gómez-Sagasti et al., 2012; Orgiazzi et al., 2016; Ritz et al., 2009). Typically, a 'biological indicator' refers to measuring the biomass, abundance, activity and/or biodiversity of common/representative species playing important roles in the ecosystem, such as earthworms, bacteria and fungi, collembola and nematodes. Thus, these biological (or ecological) indicators (i.e. *bioindicators*) can be used to assess the status and changes in ecological soil properties and processes within a given physico-chemical context, and increasingly are valued for inclusion in soil quality assessment, site-specific management strategies, measuring the state of ecosystems and for monitoring the progress of ecosystem recovery or restoration (Doran and Zeiss, 2000; Gómez-Sagasti et al., 2012; Orgiazzi et al., 2016; Ritz et al., 2009).

It has been argued by several authors that soil quality can only really be assessed in relation to one or several soil functions, ecosystem services or soil threats (i.e. relating to its 'fitness for use') (Baveye et al., 2016; Bünemann et al., 2018; Kibblewhite et al., 2008; Thomsen et al., 2012; Volchko et al., 2014b, 2013). Grouping individual, correlated SQI into higher-level categories (Figure 2-3) such as ecosystem health attributes or ecosystem services can facilitate interpretation of soil quality assessments, improve communication with stakeholders as well as provide long-term monitoring programs with the ability to adapt through time against changes in techniques, methods, interests, etc. (Burgess et al., 2018, 2017, 2016; Epelde et al., 2014a, 2014b; Garbisu et al., 2011; Gómez-Sagasti et al., 2012). Grouping of SQI according to ecosystem services has been demonstrated to be useful in, for example, evaluating aggregated microbial parameters according to a soil quality index (Burgess et al., 2017, 2016; Epelde et al., 2014b; Garbisu et al., 2011) and incorporating SQI within a set of ecosystem services as assessment endpoints in ecological risk assessment (Faber and Van Wensem, 2012; Thomsen et al., 2012).

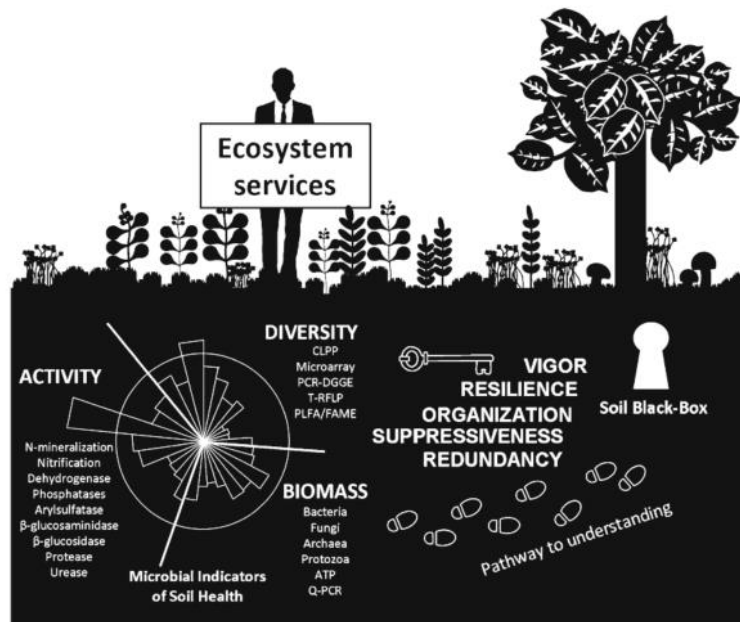


Figure 2-3. For a better interpretation of soil microbial properties as indicators of soil quality, it might be helpful to group microbial properties within a set of ecosystem health attributes of ecological relevance: vigour, organisation, resilience, suppressiveness, and redundancy, from (Gómez-Sagasti et al., 2012).

2.2 Contaminated land management

Recent high-level reports by the Food and Agriculture Organization (FAO) of the United Nations have investigated the state of knowledge of soil biodiversity (FAO et al., 2020) and assessed the global effects of soil pollution (FAO and UNEP, 2021). In these reports and others, soil contamination (used here as synonymous to pollution) has been identified as posing serious risks to human health and environmental contamination generally has been declared the largest environmental cause of disease and premature death (Landrigan et al., 2018; Science Communication Unit, University of the West of England, 2013). Environmental contamination is also considered to be one of the largest global threats to ecosystems caused by anthropogenic pressures that can affect wildlife species and ecological communities, thus driving biodiversity loss (FAO and UNEP, 2021). Many of these pressures can lead to changes in ecosystem structure and function but it can be difficult to isolate and attribute effects of contamination where many pressures are present (FAO and UNEP, 2021). In effect, soil contamination hinders the achievement of many of the Sustainable Development Goals (SDGs), including at least those related to poverty elimination (SDG 1), zero hunger (SDG 2), good health and well-being (SDG 3), protecting the most vulnerable, especially children and women (SDG 5), supplying safe drinking water (SDG 6), mitigating climate change (SDG 13), and preventing land degradation and loss of terrestrial (SDG 15) and aquatic (SDG 14) biodiversity as well as increasing the security and resilience of cities (SDG 11) (FAO and UNEP, 2021).

The direct (and indirect) effects of soil contamination on soil biota are manifold but ultimately result in an impoverishment of the soil ecosystem thus inhibiting the soil's ability to provide

key ecosystem services (FAO et al., 2020; FAO and UNEP, 2021; Orgiazzi et al., 2016; Turbé et al., 2010) (Figure 2-4). Regarding contaminated sites in particular, the microbial community structure in soil can be markedly affected by contamination, jeopardizing the provision of essential ecosystem services; thus, it is important to verify that during remediation processes, the links between soil biodiversity and soil functioning (as well as the corresponding ecosystem services) are restored (Gómez-Sagasti et al., 2012).

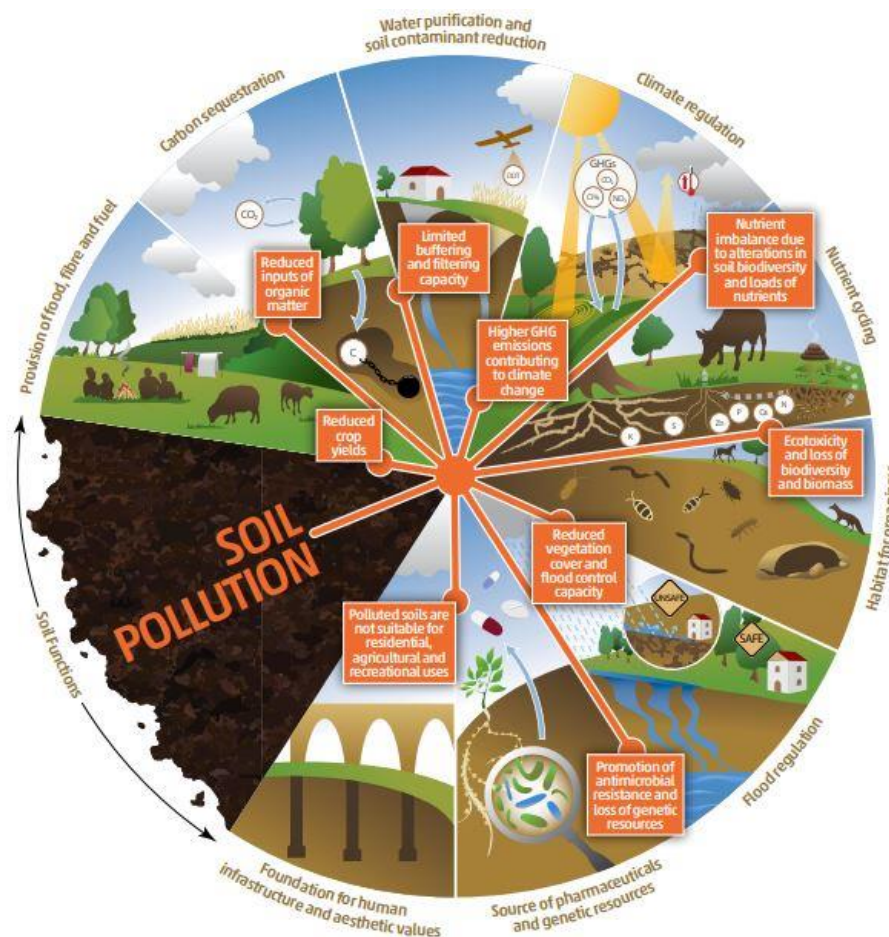


Figure 2-4. Soil contamination causes a cycle of degradation processes that leads to the reduction and ultimately to the loss of ecosystem services, from (FAO and UNEP, 2021).

Risk assessment at contaminated sites is based on the source-pathway-receptor (S-P-R) concept, also referred to as 'contaminant linkages' (UK Environment Agency, 2021). In this risk assessment framework, the mere presence of a hazard (e.g. soil contamination) does not necessarily mean that it constitutes a risk (Swartjes, 2011). For a risk to occur, there must be a source (hazard), a receptor (something that could be adversely affected) and an exposure pathway linking the source to the receptor (Bardos et al., 2020a, 2020b; Cundy et al., 2016; Swartjes, 2011). A receptor might be a human, an ecologically sensitive site, species or ecosystem, surface or groundwater resource, archaeological resource, property such as a building, crops or fisheries, or ecosystem 'goods and services' may become increasingly important receptors to consider (Bardos et al., 2020a, 2020b). Receptors, can potentially be exposed to soil contaminants through several exposure pathways, and if the risk assessment has

determined a viable exposure risk, an (eco)toxicological assessment can then be carried out to determine what adverse effects may arise depending on the estimated dosage (Swartjes, 2011).

Human health is always a protection target of vital importance when assessing risks at contaminated sites. In the Swedish EPA's (SEPA) soil guideline value model (SEPA, 2016, 2009), the following main exposure pathways are accounted for in human health risk assessment: ingestion of contaminated soil, ingestion of plants (grown on contaminated sites that may have elevated concentrations), inhalation of dust, inhalation of vapour, dermal contact and intake of drinking water (if taken from a well on site). Regarding the environment, the main protection targets accounted for in Sweden are the soil ecosystem, groundwater as a resource and surface water ecosystems. These receptors can be exposed via spreading of contaminants in free phase, porewater, etc. which is largely dependent on the specific contaminant's bioavailability (i.e. the readily available fraction of a contaminant that can cross cell membranes to enter the organism) and solubility, which in turn is heavily influenced by site-specific conditions (Naidu et al., 2015; SEPA, 2016; Swartjes, 2011). Fully understanding the actual risks posed by contaminants to sensitive receptors requires a more complex, site-specific risk assessment wherein a critical factor is the bioavailability of contaminants (Naidu et al., 2015; Swartjes, 2011).

Risk management interventions to mitigate/reduce the risks can take place at any point in the S-P-R chain as long as it breaks the contaminant linkage, which could involve removing the source, disrupting the pathway or managing the receptor to reduce the risk of unacceptable harm (Bardos et al., 2020a, 2020b; Cundy et al., 2016; Swartjes, 2011). A variety of remediation options are available that target different points across the various contaminant linkages. Conventional soil remediation techniques are those that utilise physical, chemical, biological or a combination of methods to, most often, address the source of contamination ex-situ (entailing soil excavation and subsequent treatment on- or off-site via soil washing, thermal treatment, etc.) or in-situ to degrade, transform, extract or stabilise (in)organic contaminants at the site or utilise barriers like clay liners and permeable reactive barriers to isolate the site from its surroundings (Kuppusamy et al., 2016b, 2016a; Swartjes, 2011). The current international consensus is that land contamination decision making should be made on the basis of risks to human health and the wider environment, according to S-P-R linkages, a paradigm often referred to as risk-based land management (RBLM) (Bardos et al., 2020b, 2018; P. Bardos et al., 2011; Vegter et al., 2002). RBLM provides an objective way to link actions to the prevention of harm, a rationale for how to intervene (i.e. managing contaminant linkages), and a rationale to prioritise the dispensation of limited resources at sites according to risk evaluation (Bardos et al., 2020a, 2020b, 2018; P. Bardos et al., 2011; Common Forum and NICOLE, 2013; Swartjes, 2011; Vegter et al., 2002). At its core, RBLM is predicated on the reduction of risks to human health and the environment to the degree necessary to ensure a safe, beneficial reuse of site (i.e. fitness for use) while protecting the environment over the long-term (Bardos et al., 2020a, 2018; ISO, 2017; Swartjes, 2011). It can, however, encounter challenges to enmesh with existing environmental objectives and gain acceptance from regulatory agencies due to the emphasis on full decontamination and source removal (Swartjes, 2011).

Remediation is not inherently sustainable and poorly planned projects, or high-impact remediation options, can have substantial negative effects (Anderson et al., 2018; Bardos et al., 2020a; Cundy et al., 2016). This is a common issue with many remediation techniques, especially ex-situ measures but also some in-situ techniques, which can also result in the soil ecosystem and its essential functions being seriously degraded or even eliminated by the remedial action (Gerhardt et al., 2017; Swartjes, 2011). Consequently, contaminated soil has often been viewed as waste to be disposed of rather than as a valuable resource to be treated and reused (Gerhardt et al., 2017; Mench et al., 2010). In recent years, sustainable remediation frameworks have been adopted to reduce the economic, social and environmental costs of remediation and to ensure that a brownfield redevelopment project is more broadly beneficial (a net benefit) to society (Bardos, 2014; Bardos et al., 2020b, 2018). Sustainable remediation has been described as *'the practice of demonstrating, in terms of environmental, economic and social indicators, that the benefit of undertaking remediation is greater than its impact and that the optimum remediation solution is selected through the use of a balanced decision-making process'* (Bardos, 2014; Bardos et al., 2018). Definitions vary and similar concepts with varying degrees of strong/weak sustainability exist, but sustainable remediation as a concept most often refers to the use of wide-ranging indicators, typically as measurable endpoints, to evaluate the sustainability of a remediation project according to environmental, economic and social criteria that align with sustainable development principles (Anderson et al., 2018; Bardos, 2014; Bardos et al., 2020a; P. Bardos et al., 2011; Harwell et al., 2021; ISO, 2017; Smith, 2019). Sustainable remediation and sustainable brownfield regeneration can be seen as overlapping domains in the wider context of sustainable land development (ISO, 2017; Rizzo et al., 2016). Therefore, risk management should also meet sustainable development principles as a core project objective, and this integrated approach constitutes sustainable risk-based land management (SRBLM) (Bardos et al., 2020a, 2020b; Common Forum and NICOLE, 2013; Rizzo et al., 2016). SRBLM has emerged as the optimal approach for balanced contaminated land decision-making, which combines a risk-based framework for determining when the risk (or potential risk) is unacceptable and where/when action is necessary with ensuring that sustainability is a part of deciding how such unacceptable risks are to be managed (Bardos et al., 2020b). It aims to ensure that a balanced decision is taken which optimizes overall benefit and achieves the best solutions to manage risks at contaminated sites (Bardos et al., 2020b; Common Forum and NICOLE, 2013).

Increasingly, new trends in sustainable remediation (e.g. land stewardship (Common Forum and NICOLE, 2018)) call for accounting for soil (even if contaminated) as a valuable resource that can be cleaned and made fit for 'soft' end uses like green spaces, which require ecological functioning (Bardos et al., 2016; Cundy et al., 2016; Menger et al., 2013; Volchko et al., 2014a). As previously discussed in Drenning et al. (2020) (Paper I), the benefits offered by soil functioning at its true capacity are also essential to achieve environmental goals such as the Sustainable Development Goals (SDGs), many of which are ultimately dependent on a healthy, thriving soil biodiversity (FAO et al., 2020; FAO and UNEP, 2021; Orgiazzi et al., 2016). Many of the SDGs also have a strong connection to land and water management, which has been

highlighted by Keesstra et al. (2018a, 2016) and in the Land Stewardship approach by linking to the SDGs (Common Forum and NICOLE, 2018).

2.3 Gentle remediation options (GRO)

Vegetation-covered urban brownfields are important, but underappreciated, elements of urban green infrastructure (Mathey et al., 2018, 2015), and gentle remediation options (GRO) are NBS that can be applied to manage risks at brownfields and provide or maintain vital ecosystem services (Bardos et al., 2020a, 2016; Cundy et al., 2016; Song et al., 2019). GRO are defined as *risk management strategies or technologies that result in a net gain (or at least no gross reduction) in soil function as well as achieving effective risk management* (Cundy et al. 2016). GRO is an umbrella term covering a set of remediation technologies based upon the use of plant (phyto-), fungi (myco-), and/or bacteria-based (bio-) methods with or without the use of chemical additives or soil amendments (Table 2-2) (Cundy et al., 2016; GREENLAND, 2014a). Soil invertebrates such as earthworms (vermi-) have also been shown to improve decontamination of organic (e.g. pesticides) and inorganic contaminants (metals) by plants and microorganisms (FAO et al., 2020; Lacalle et al., 2020; Orgiazzi et al., 2016; Rodriguez-Campos et al., 2014; Turbé et al., 2010), and could also be considered a GRO.

Table 2-2. List of definitions for GROs used to remediate soils contaminated by either trace elements or mixed contamination, adapted from (Bardos et al., 2020a; Cundy et al., 2016; GREENLAND, 2014a; OVAM, 2019).

GRO	Definition
Phytoextraction	Process in which plants and their associated microorganisms absorb contaminants and fix them in above-ground plant tissue that can then be removed from the site during harvesting.
Phytodegradation/ phytotransformation	The use of plants (and associated microorganisms like endophytic bacteria) to uptake, store and degrade contaminants.
Rhizodegradation	The use of plant enzymes and rhizospheric (in root zone) microorganisms to degrade organic contaminants.
Phytostabilisation	Reduction in the bioavailability and mobility of contaminants by immobilisation in root systems and/or living dead biomass in the rhizosphere soil.
Phytovolatilisation	The use of plants to remove contaminants from the growth matrix, transform them to less toxic forms and disperse them (or their degradation products) into the atmosphere.
In-situ immobilisation	Reduction in the bioavailability of contaminants by immobilisation or binding them to the soil matrix through the incorporation into the soil of organic or inorganic compounds to prevent excessive uptake and transfer into the food chain.
Phytoexclusion	The implementation of a stable vegetation cover using excluder plants which do not accumulate contaminants in the harvestable biomass, often combined with in-situ immobilisation.
Rhizofiltration	The removal of contaminants from aqueous sources by plant roots and associated microorganisms.
Phytohydraulics	Process in which plants and their microorganisms take up and evaporate water and thereby influence the groundwater level, the direction and velocity of the groundwater flow.
Bioremediation	Generic term applied to a range of remediation and risk management technologies which utilise soil microorganisms to degrade, stabilise or reduce the bioavailability of contaminants.
Mycoremediation	A form of bioremediation in which fungi-based methods are used to degrade, extract, stabilise or reduce the bioavailability of contaminants.
Vermiremediation	A remediation technique which utilises earthworms to remove or stabilise soil contaminants.

These more innovative biological methods of soil remediation have emerged as alternatives to conventional physicochemical methods, which may be unsuitable or unnecessary in many cases, and to provide multi-functionality for: i) effective risk management, ii) a reduction of

soil ecotoxicity, iii) the legal and ethically required reduction of contamination risks for both human health and the environment; and, concurrently, a recovery of iv) soil health and v) associated ecosystem services (Burges et al., 2018; Cundy et al., 2016; GREENLAND, 2014a; Lacalle et al., 2020). Substantial economic (e.g. biomass generation), socio-cultural (e.g. leisure and recreation), and environmental (e.g. ecosystem services and restoration of plant and microbial and animal communities) co-benefits are also possible through GRO application when intelligently applied (Bardos et al., 2016; Conesa et al., 2012; Cundy et al., 2016, 2013a; Evangelou et al., 2012; GREENLAND, 2014a).

In terms of risk management, GRO are primarily applied on contaminated soils to reduce contaminant transfer to local receptors by gradually removing the bioavailable pool of inorganic contaminants (*phytoextraction*), removing or degrading organic contaminants (*phyto- and rhizodegradation*), filtering contaminants from surface water and waste water (*rhizofiltration*) or groundwater (*phytohydraulics*), and stabilising or immobilising contaminants in the soil matrix (*phytostabilisation, in-situ immobilisation*) often in combination with vegetation cover using excluder plants (*phytoexclusion*) (Table 2-2). If well-designed, GRO can be customised to provide risk management along S-P-R contaminant linkages via i) gradual removal or immobilisation (i.e. reducing bioavailability/solubility) of the contaminant source, ii) managing the flux of contaminants along exposure pathways and breaking connections to receptors through containment and stabilisation, and iii) managing the receptor's access to the contaminated medium thus preventing exposure (Bardos et al., 2020a; Cundy et al., 2016; GREENLAND, 2014a). While GRO may not be well-suited to highly contaminated sites, hotspots or point source terms such as buried tanks or oil spills, they are particularly suitable for large areas and contaminated sites that pose low to medium risks to human health and the environment (Andersson-Sköld et al., 2014; Cundy et al., 2016; Enell et al., 2016; GREENLAND, 2014a). GRO are useful as 'primary prevention strategies' in various applications to reduce or eliminate human (and non-human) exposure to contaminants (Henry et al., 2013). GRO can also be used for source removal of inorganic and organic contaminants though the timeframe for remediation can differ significantly between the contaminants and the mechanisms involved (Figure 2-5). An important note is that the 'relative remediation time' as used in Figure 2-5 represents only the estimated time it would take for full source removal (e.g. via extraction or degradation) and can vary depending on if total or bioavailable concentrations are used as a benchmark.

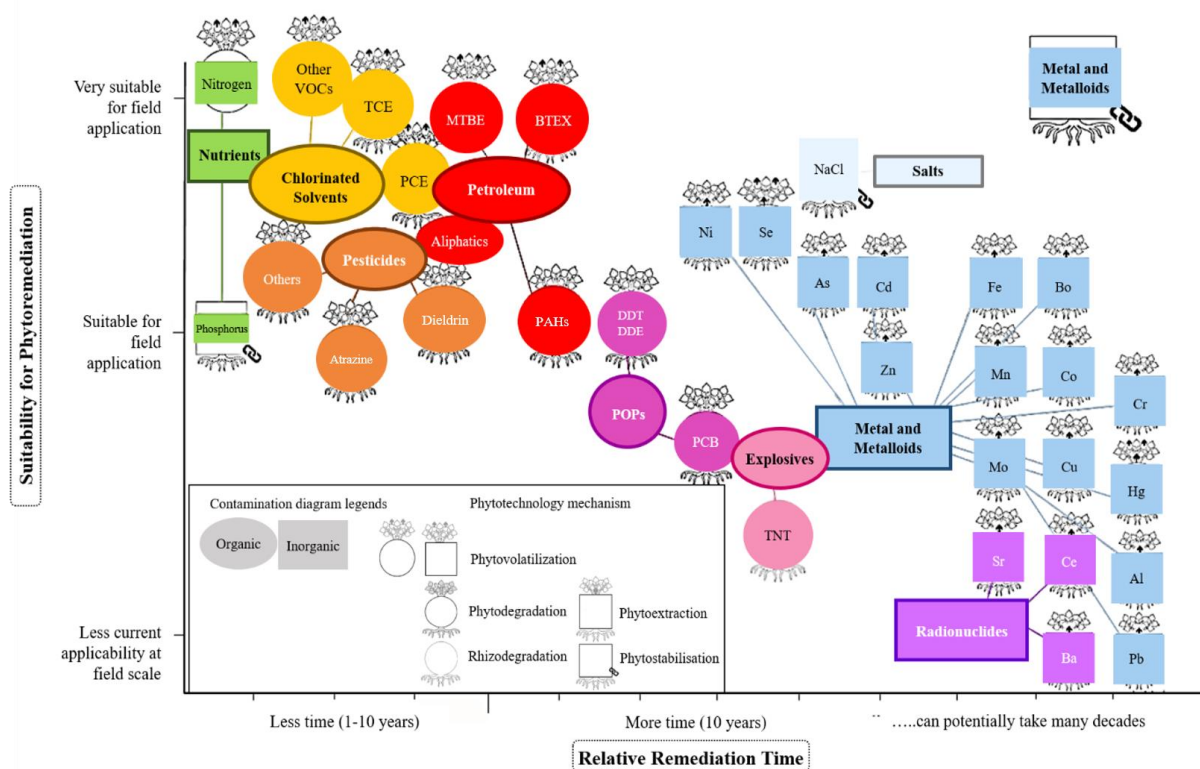


Figure 2-5. Relative remediation time for source removal (relevant only for extraction, degradation and volatilisation) of groups of contaminants and applicability of the phytotechnology mechanisms. Colours correspond to contaminant grouping. After (OVAM, 2019) and (Kennen and Kirkwood, 2015).

In practice, many of the GRO techniques can be separated into 'standard' and 'enhanced' phytoremediation. Standard refers to using the inherent functions of plants and their naturally occurring microbes that enable the various mechanisms. To improve the effectiveness of GRO, phytoremediation can be enhanced (or 'aided' or 'microorganism-assisted') through enriching the microbes in the rhizosphere or within the plant itself by bioaugmentation (i.e. introducing external species to the site that may be better suited for degrading specific contaminants) or biostimulation (i.e. enhancing the already existing microbes by the use of soil amendments) that can promote plant growth and tolerance and increase degradation and extraction rates (Mench et al., 2010; OVAM, 2019; Thijs et al., 2017, 2016; Vangronsveld et al., 2009).

The following sections describe each of the GRO techniques in brief detail; for more information and compilations of field studies the reader is referred to Drenning (2021a) and other more extensive, in-depth reports, e.g. (Gerhardt et al., 2017, 2009; GREENLAND, 2014a, 2014b; Mench et al., 2019, 2010; Moreira et al., 2021, 2019; Vangronsveld et al., 2009). A summary table compiling GRO mechanisms, contaminants and media for which they are applicable and possible plant species are presented in Table 4-3.

2.3.1 Gentle remediation of organics

Gentle remediation of organics by degradation aims at the complete mineralisation of organic contaminants into carbon dioxide, nitrate, chlorine, ammonia and other elemental constituents

of the initial molecule (Mench et al., 2010; OVAM, 2019). This remediation strategy has been proven viable for a wide variety of organic compounds, including 1) petroleum products – polycyclic aromatic hydrocarbons (PAHs), aliphatic hydrocarbons, fuels and BTEX compounds; 2) persistent organic pollutants – polychlorinated biphenyls (PCBs), DDT and other pesticides; 3) explosives – nitro-aromatics such as trinitrotoluene (TNT); and 4) chlorinated solvents – linear halogenated hydrocarbons such as trichlorethylene (TCE) (Gerhardt et al., 2017, 2009; Kennen and Kirkwood, 2015; Mench et al., 2010; OVAM, 2019). In general, plants are utilised for the gentle remediation of organics in two primary ways: 1) to speed up the natural attenuation process (biodegradation) – plant natural biological process can facilitate biodegradation by, for example, improving microbial activity by supplying a variety of root exudates into the root zone that create the conditions for a rich microbiome as well as potentially inducing the breakdown of organic compounds by specific bacteria, plant roots growing and extending through the soil to spread microbes throughout the soil profile, and functioning as 'bio-pumps' by transpiring water and supplying oxygen to microbes; and 2) to control, degrade and volatilise organic contaminants in groundwater (hydraulic control) – in their function as 'bio-pumps', deep-rooted plants with high evapotranspiration rates that take up large amounts of water (e.g. phreatophyte trees) can hydraulically control the direction, velocity and flux of contaminants in contaminated groundwater plumes to prevent spreading off-site (Gerhardt et al., 2017, 2009; Jambon et al., 2018; Kennen and Kirkwood, 2015; Mench et al., 2010; OVAM, 2019; Robinson et al., 2006; Thijs et al., 2017, 2016).

Phytodegradation refers to the process of plant uptake of contaminants and consequent degradation in the plant by metabolic processes or enzymes secreted by the plant or microorganisms (e.g. dehalogenases, nitro-reductases, oxophytodienoate reductases, polyphenol oxidases, peroxidases, laccases, dehydrogenases, hydrolases) (Gerhardt et al., 2017, 2009; OVAM, 2019; Wolfe and Hoehamer, 2003). Plant uptake is dependent on the bioavailability of the contaminants, and the hydrophobicity of the organic compounds is a major determining factor, for which a $\log K_{ow}$ value between 0.5 – 3.5 indicates a good uptake by plants. Organic compounds amenable to plant uptake include various chlorinated solvents, herbicides, pesticides, insecticides, explosives and low molecular weight petroleum products (OVAM, 2019). Effectiveness of *in-planta* degradation is highly dependent on the plant's tolerance to potential toxicity and the microorganisms' capacity to break down contaminants.

Rhizodegradation entails similar degradation processes but refers specifically to degradation occurring in the rhizosphere through microbial activity (OVAM, 2019). Many organic compounds are too hydrophobic to be taken up by plants (e.g. PAHs, PCBs) but can be degraded outside the plant, which occurs to some extent even if a compound is absorbed into the plant (OVAM, 2019). Certain plant species have been identified as being more effective for remediating specific organic contaminants due to their ability to 'selectively recruit', a function of their specific root exudates, bacterial and fungal communities to the rhizosphere that are both tolerant to the contamination as well as able to break down the contaminants present in the soil (Jambon et al., 2018; Mench et al., 2010; Thijs et al., 2017, 2016).

Phytovolatilisation refers to the process by which volatile contaminants are excreted from the leaves of plants by evapotranspiration (OVAM, 2019). Plants can take up certain hydrophilic

organic compounds (i.e. $\log K_{ow}$ between 0.5 – 3.5) and metabolise them into different, less toxic forms for sequestration; however, some plants preferentially release volatile contaminants (e.g. TCE and BTEX compounds) by transpiration via the leaves (Kennen and Kirkwood, 2015; McCutcheon and Schnoor, 2003; Schnoor, 1997). Phytovolatilisation can indeed remove certain contaminants from soil, but it can also simply shift the problem to another environmental compartment (i.e. air) and contaminants may eventually be redeposited to the soil downstream of the site by precipitation (Gerhardt et al., 2017; OVAM, 2019; Vangronsveld et al., 2009). Even if a contaminant has been converted to less toxic form, its release means it is still in the environment though it may be diluted in the atmosphere to such low levels that it poses insubstantial risks (Gerhardt et al., 2017) or is degraded by photolysis (i.e. UV and hydroxyl radicals in the atmosphere). Preferably, microorganisms can be used to enhance the degradation processes so that degradation occurs within the plant or rhizosphere (OVAM, 2019; Vangronsveld et al., 2009). For instance, specific, customised endophytic bacteria have been successfully inoculated into plants to greatly reduce or eliminate altogether the volatilisation of TCE and BTEX compounds by enhancing degradation thereby decreasing phytotoxicity and evapotranspiration (OVAM, 2019; Vangronsveld et al., 2009; Weyens et al., 2009a, 2009b).

2.3.2 Gentle remediation of inorganics and persistent organics

Gentle remediation of inorganics can aim to mitigate risks by either 1) gradually removing the source of the contamination by harvesting plants that have accumulated the contaminants, or 2) managing the exposure pathways by reducing the spreading of contaminants in porewater, groundwater or the atmosphere (Mench et al., 2010; OVAM, 2019; Robinson et al., 2006; Vangronsveld et al., 2009). These two strategies are predominantly applied to manage metal(loid)s, including As, Cd, Cu, Cr, Hg, Ni, Pb, Zn, etc., as well as salts, excess nutrients, radionuclides and even certain persistent organic contaminants like DDT and PCBs (Gerhardt et al., 2017; Kennen and Kirkwood, 2015; Mench et al., 2010; OVAM, 2019; Vangronsveld et al., 2009)

Phytoextraction is arguably the most well-known and thoroughly tested GRO. The aim is to remove the source of contamination by utilising the capacity of plants to function as 'bio-pumps' to take up contaminants from soil and groundwater into their biomass, which can then be removed from the site by harvesting the plant (Mench et al., 2010; Robinson et al., 2009, 2006, 2003a; Vangronsveld et al., 2009). There are three basic phytoextraction strategies common in practice: 1) continuous or natural phytoextraction using hyperaccumulators (e.g. *Noccaea caerulescens* for Cd and Zn, *Pteris vittate* for As); 2) continuous or natural phytoextraction of trace elements using fast-growing, high biomass producing plants (e.g., *Salix* or *Populus* spp., sunflower, tobacco, *Brassica* spp., maize, wheat, grasses); 3) enhanced or chemically-assisted phytoextraction by using soil amendments (e.g., chelators or acidifying amendments) to increase trace element mobility in the soil for greater uptake in plants, though this can increase the risk of leaching (GREENLAND, 2014a; Vangronsveld et al., 2009). The maximum uptake in all these approaches depends on two main variables: i) contaminant concentration in harvestable plant parts, which can be estimated using the bio-accumulation (or bio-concentration) factor (BAF/BCF), and ii) harvestable biomass yield since high biomass

production results in greater amounts of contaminants being removed (Burgess et al., 2018; Keller, 2005; Keller et al., 2003; Robinson et al., 2015, 2006; Vangronsveld et al., 2009).

Phytostabilisation is an alternative strategy to source removal where contaminants remain on site and plants (often 'aided' with soil amendments) are instead utilised to reduce the mobility and bioavailability of contaminants in the environment thus mitigating adverse effects (Cundy et al., 2016; Epelde et al., 2009b; Gerhardt et al., 2017; Mench et al., 2010; OVAM, 2019; Vangronsveld et al., 2009). Phytostabilisation is predominantly applied to immobilise metal(loid)s in soil but it can also be useful for capturing recalcitrant organic compounds like PAHs, DDT and PCBs that can then be degraded over time via rhizodegradation (OVAM, 2019). More specifically, phytostabilisation uses plants and their associated microbes for long-term containment of contaminants such as metals in solid matrices through adsorption, absorption and accumulation in the roots, precipitation in the root zone or by physical stabilisation of the soil that either prevents or minimises their mobility in the food chain, downward percolation to groundwater and re-entrainment of contaminated particulates for direct inhalation or ingestion by humans (Cundy et al., 2016; Epelde et al., 2009b; Gerhardt et al., 2017; Mench et al., 2010; OVAM, 2019; Vangronsveld et al., 2009). One of the main features of phytostabilisation is improving the quality of the soil to enable the revegetation of contaminated, derelict brownfield sites, which, with its associated natural attenuation mechanisms, is recognised to be the most realistic remedial action to reduce the risks of exposure to receptors at many of these sites (Dickinson et al., 2009). Another key aspect of (aided) phytostabilisation is that it can both reduce the toxicity of contaminants as well as improve ecosystem functioning by restoring soil health and increasing microbial activity, biomass and diversity in the long-term (Burgess et al., 2018; Epelde et al., 2009b; Kumpiene et al., 2009; Touceda-González et al., 2017a, 2017b). Indeed, the demonstration of the recovery soil functionality and soil health at contaminated sites might be a key factor to increase the acceptance of phytostabilisation as a viable remediation option (Epelde et al., 2009b; Kumpiene et al., 2009; Vangronsveld et al., 2009).

Many plants have been demonstrated to exclude certain contaminants (i.e. **phytoexclusion**) by avoiding uptake altogether, immobilising them in the roots or restricting uptake to the shoots to avoid sensitive organelles. Annual crops that exclude specific contaminants for uptake, particularly Cd, are highly valued for use on agricultural soils where contaminant transfer into the food chain is a risk (Dickinson et al., 2009; GREENLAND, 2014b; Haller and Jonsson, 2020; Kidd et al., 2015; Tang et al., 2012). Many staple crops like cereals and vegetables have been shown to have the ability to either exclude toxic metals like Cd from uptake or translocate only miniscule amounts to their harvestable, edible biomass, including certain species/cultivars of wheat (*Triticum* spp.), barley (*Hordeum vulgare*), rice (*Oryza sativa*), potato (*Solanum tuberosum*), soybean (*Glycine max*) and maize (*Zea mays*). The use of metal-excluding cultivars of annual crops can be an effective option for mitigating the risk of contaminant transfer into the food chain on agricultural land (Andersson-Sköld et al., 2013a; Dickinson et al., 2009; GREENLAND, 2014b; Kidd et al., 2015; Tang et al., 2012).

2.3.3 Soil amendments

The abovementioned GRO techniques are often used in combination with soil amendments (i.e. 'aided') to enhance effectiveness by reducing (or increasing) the bioavailability of metals in soil and uptake in plants as well improve soil quality, particularly when using organic amendments, to enable the establishment of vegetation in poor soils by, for example, improving soil physical properties like bulk density and pore structure, improving water infiltration and holding capacity, improving soil fertility by adding essential micro- and macronutrients, balancing soil pH, re-establishing microbial communities and increasing soil organic matter (Burgess et al., 2018; Epelde et al., 2009b; Gómez-Sagasti et al., 2018; GREENLAND, 2014b; Kidd et al., 2015; Kumpiene et al., 2019; Mench et al., 2010; Vangronsveld et al., 2009). The positive effects can also be compounded through the use of effective agronomic techniques (Kidd et al., 2015). Use of organic and inorganic amendments could also enable the recycling of wastes, residues and diverse by-products to promote a circular economy (Chowdhury et al., 2020; Gómez-Sagasti et al., 2018; Lacalle et al., 2020; Míguez et al., 2020).

When soil amendments are used independently to immobilise contaminants, it is referred to ***in-situ* (chemical) immobilisation** (Dickinson et al., 2009; GREENLAND, 2014b; Kidd et al., 2015; Kumpiene et al., 2019, 2008; Mench et al., 2010; Vangronsveld et al., 2009). Whether to mobilise or immobilise contaminants by manipulating their bioavailability in soil, especially in the case of inorganics, is a key factor to consider when deciding upon a GRO strategy based on extraction or stabilisation, respectively, and various soil amendments can be used to achieve different aims (Bolan et al., 2014; Vangronsveld et al., 2009). Soil amendments commonly used for *in-situ* immobilisation and aided phytostabilisation of trace elements can be broadly broken down into inorganic and organic amendments (GREENLAND, 2014b):

1. **Inorganic** – rock phosphate (a major source of P fertilisers), Thomas basic slag (a by-product of the iron industries), wood ashes, cyclonic ashes, zerovalent iron grit, Linz-Donawitz slag, siderite, gravel sludge, red mud, drinking water residues
2. **Organic** – animal manures and slurries, biosolids (sewage sludge), composted biosolids, green waste composts, biochar

Following an extensive review of *in-situ* immobilisation (including aided phytostabilisation), Kumpiene et al. (2019) conclude that most of the field studies implementing these techniques show a certain degree of improvement in the soil and/or vegetation status following soil amendment. As soil toxicity decreases, plants and microorganisms will colonize the treated soil, which will induce dissolution/precipitation reactions and drive the geochemical soil conditions away from equilibrium, but the net effects of such processes on trace elements circulation can only be evaluated by monitoring the sites over extended time periods (Kumpiene et al., 2019).

2.3.4 Vegetation cover

One of the main features of plant-based GRO strategies is improving the quality of the soil to enable the revegetation of contaminated, derelict brownfield sites (Dickinson et al., 2009). A **vegetation cover**, including resultant root growth and exudates, may also produce beneficial changes in soil parameters that improve soil aggregation and binding of contaminants by stimulating soil biota and providing litter through leaf fall (Dickinson et al., 2009). Establishing

a vegetation cover can also be a suitable primary exposure prevention strategy at sites where source removal by GRO (or other conventional alternatives) is not possible due to constraints imposed by phytotoxicity, budgetary limitations, timescale, risks due to grazing livestock and/or if there is not alternative treatment easily available (Mench et al., 2010). Also, in many cases, contaminated topsoil is left barren or with sparse vegetation thus prone to spreading contaminants off-site by wind erosion as dust emission, water erosion via stormwater runoff into local surface water or by leaching into groundwater (Burgess et al., 2018; Cundy et al., 2016; Dickinson et al., 2009; Gerhardt et al., 2017; Mench et al., 2010; OVAM, 2019; Vangronsveld et al., 2009). Barren or sparsely vegetated brownfields can pose significant human health risks due to inhalation of dust-borne contaminants, which can be the most significant human health risk at some sites (Dickinson et al., 2009; Gil-Loaiza et al., 2018; Monica O. Mendez and Maier, 2008).

The vegetation cover created in plant-based GRO offers valuable secondary effects that can provide effective risk management, including: 1) **erosion control** by physically stabilising the soil with fibrous root networks, increasing soil porosity and extensive canopy cover to reduce runoff and prevent horizontal and lateral migration (GREENLAND, 2014a; ITRC, 2009; Kennen and Kirkwood, 2015; Mench et al., 2010; OVAM, 2019; Vangronsveld et al., 2009).; 2) **hydraulic control** by both influencing the flow and direction of groundwater and reducing the flux of contaminants (i.e. lateral spreading or vertical leaching and mobilisation) to groundwater via plants acting as 'bio-pumps', especially those with high rates of evapotranspiration (Barac et al., 2009; Ferro et al., 2013; GREENLAND, 2014a; ITRC, 2009; Kennen and Kirkwood, 2015; Mench et al., 2010; OVAM, 2019; Pivetz, 2001; Robinson et al., 2003a; Vangronsveld et al., 2009); 3) **dust control** by greatly reducing the total dust flux and emission of fine particulates mobilised by wind, including PM1, PM2.5 and PM4 (i.e. particulate matter of 1, 2.5 and 4 μm diameter respectively) which represent the greatest health risks and potential for long-distance transport (Cundy et al., 2016; Gil-Loaiza et al., 2018; GREENLAND, 2014a; Henry et al., 2013; Monica O. Mendez and Maier, 2008). Especially in urban areas, vegetation can further improve air quality by filtering and capturing airborne contaminants (e.g. PCBs) as they adhere to the waxy cuticle of plant leaves and bark (Henry et al., 2013; Kennen and Kirkwood, 2015). 4) Vegetation can also function as a natural barrier between humans and the contaminated soil to '**manage receptor access**' and mitigate exposure by soil ingestion or dermal contact (Bert et al., 2012; Cundy et al., 2016; GREENLAND, 2014a; Kidd et al., 2015).

2.3.5 Gentle remediation in aqueous media

GRO can be applied to manage various contaminated media, including groundwater, surface water and wastewater. **Rhizofiltration** refers to the use of plants to protect surface water resources through the continuous removal of contaminant solutes in aqueous media by accumulation into or adsorption onto plant roots as well as degradation by associated microorganisms (GREENLAND, 2014a; Kennen and Kirkwood, 2015; McCutcheon and Schnoor, 2003; OVAM, 2019; Pivetz, 2001). Most often, the term rhizofiltration is used to describe application of vegetation to filter contaminants from surface water as e.g. constructed wetlands, wastewater irrigation or stormwater biofilters (Kennen and Kirkwood, 2015;

McCutcheon and Schnoor, 2003; OVAM, 2019; Pivet, 2001). Rhizofiltration is essentially the key mechanism underlying much of 'Low Impact Development' (LID) and 'Sustainable Urban Drainage Systems' (SUDS), which have become key features or 'Best Management Practices' (BMP) for sustainable urban stormwater management and green infrastructure (Cundy et al., 2016; Kennen and Kirkwood, 2015; Menger et al., 2013). GRO may be particularly valuable in combination with urban flood management strategies by intercepting and delaying stormwater runoff, surface and groundwater flow management, reducing contaminant transfer to water bodies, soil erosion prevention, and by increasing permeable surface area for greater infiltration (Cundy et al., 2016; Kennen and Kirkwood, 2015; Menger et al., 2013). Many applications show that rhizofiltration systems can also provide effective, continuous surface and wastewater treatment (ANL, 2008; Cundy et al., 2020; Dimitriou and Aronsson, 2005; Kennen and Kirkwood, 2015; Marchand et al., 2010; Pivet, 2001). Either aquatic (e.g. macrophytes), coastal or terrestrial plants can be used for rhizofiltration; for example, *Phragmites australis* (common reed) and *Typha* spp. (reed, cattail) are basic species for use in constructed wetlands that are highly tolerant to a range of contaminants and high salinity (Gawronski et al., 2011; McCutcheon and Schnoor, 2003; Pivet, 2001).

Phytohydraulics is a term that describes the management of contaminants present in groundwater (ITRC, 2009; Kennen and Kirkwood, 2015; OVAM, 2019). It is based on the capacity of plants to root into groundwater aquifers and transpire sufficient amounts of water to influence the flow and direction of groundwater as well as the flux of contaminants into groundwater bodies (i.e. hydraulic control) (OVAM, 2019). Typical application would entail planting trees as a barrier to contain a contaminated groundwater plume and limit the spread of contamination, or functioning as a groundwater 'bio-pump' treatment system (Kennen and Kirkwood, 2015; OVAM, 2019). The specific term 'phytohydraulics' may not be common jargon, however there are numerous examples of trees being used for hydraulic control of contaminated groundwater plumes (e.g. (Barac et al., 2009; Cundy et al., 2020; El-Gendy et al., 2009; Ferro et al., 2013; Hong et al., 2001; Kennen and Kirkwood, 2015; Pivet, 2001)). The most suitable plant species for phytohydraulics are phreatophytes, which are deep-rooting to reach groundwater (up to 10 meters), transpire large amounts of water, prefer wet soils and can tolerate water saturated conditions (Kennen and Kirkwood, 2015; OVAM, 2019). The most prominent examples are tree species like willow (*Salix* spp.) and poplar (*Populus* spp.), but also include other deep-rooted trees such as alder (*Alnus* spp.), ash (*Fraxinus* spp.) and oak (*Quercus* spp.) and tap-rooted, herbaceous species like alfalfa (*Medicago sativa*) or many grass species accustomed to surviving drought or water-scarce conditions as in deserts or prairies (Kennen and Kirkwood, 2015; McCutcheon and Schnoor, 2003; OVAM, 2019).

2.3.6 Bioremediation

Bioremediation is a broad umbrella term that refers to the use of bacteria and/or fungi to remediate contaminated sites, primarily regarding organic contaminants such as mineral oils, petroleum hydrocarbons, PAHs, PCBs, pesticides, chlorinated solvents, etc. Bioremediation utilises microbial activity to biodegrade available contaminants within impacted ecosystems and is particularly effective for groundwater treatment although this is dependent on a number of factors such as geochemical and hydrological conditions, nutrient availability and growth

substrates, the nature and bioavailability of contaminant, the abundance and activity of microorganisms, and aerobic or anaerobic degradation pathways amongst others (FAO et al., 2020; Fingerman and Nagabhushanam, 2019; Garg et al., 2017; Haritash and Kaushik, 2009; Kuppusamy et al., 2016b; Megharaj et al., 2011; Megharaj and Naidu, 2017; US EPA, 2006). In general there are three main approaches for the bioremediation of contaminated sites: 1) monitored natural attenuation (or natural source zone depletion, NSZD) – natural decontamination processes are carried out by native microbes at the site, which is left undisturbed but monitored; 2) bioaugmentation – selected microbial strains that possess greater capacity to degrade the target contaminants at a faster rate are injected into the soil or groundwater; and 3) biostimulation – existing microbes present at site are stimulated by modifying the environment (e.g. moisture, pH, nutrients, oxygen) with various amendments to enhance biodegradation of target contaminants. Bioremediation techniques have been used in a variety of applications and have often achieved significant contaminant reduction, see e.g. (Cristaldi et al., 2017; Fingerman and Nagabhushanam, 2019; Haritash and Kaushik, 2009; Kuppusamy et al., 2016b; Lacalle et al., 2020; Megharaj et al., 2011; Megharaj and Naidu, 2017; US EPA, 2006) for more thorough reviews and information.

Mycoremediation entails using fungi directly for remediation, though the term is uncommon, and the technique is usually included within the much broader umbrella term of bioremediation. Fungi have proven to be useful for effectively remediating a wide variety of contaminants by biodegradation, biosorption and bioconversion; including heavy metals, persistent organic pollutants, textile dyes, chlorinated solvents, PAHs and other petroleum products, pharmaceuticals, pesticides, herbicides and insecticides (see e.g. (Akhtar and Mannan, 2020; Deshmukh et al., 2016; Kulshreshtha et al., 2014) for reviews). Regarding organic contaminants, certain types of fungi can, for example, enhance degradation of PAHs (Haritash and Kaushik, 2009), TNT and other explosives (Koehler et al., 2002), DDT compounds (Purnomo et al., 2011, 2010), and PCBs (Stella et al., 2017).

As a remediation technique, **vermioremediation** refers to the use of earthworms for the removal of contaminants from soil. Soil invertebrates such as earthworms have also been shown to improve decontamination of organic (e.g. pesticides) and inorganic contaminants (metals) by plants and microorganisms (FAO et al., 2020; Lacalle et al., 2020; Orgiazzi et al., 2016; Rodriguez-Campos et al., 2014; Turbé et al., 2010). Earthworms and other soil fauna ('ecosystem engineers') can also function as dispersal agents for both microorganisms that degrade organic contaminants and the contaminants themselves through the soil profile (FAO et al., 2020). In general, as earthworms burrow through soil they mix and alter the physico-chemical and biological properties of the soil by i) increasing availability of nutrients like C and N; ii) ingesting and mixing the soil with organic material; iii) affecting soil structure, pore space and aeration through burrowing; and iv) changing the soil bacterial and fungal communities by modifying the structure and size of soil aggregates. All of which can result in increased soil enzyme production and microbial activity and greater interaction with contaminants by increasing bioavailability thus resulting in enhanced biodegradation. The 'vermicasts' left behind as earthworms excrete soils are carbon- and microbe-rich and, besides

improving soil quality, can also bind contaminants to stabilise them in the soil matrix (Lacalle et al., 2020; Rodriguez-Campos et al., 2014; Sinha et al., 2008; Zeb et al., 2020).

While effective as remediation techniques, bioremediation strategies do not provide the same landscape and visual amenity, nor ecosystem service benefits possible from other nature-based solutions that utilise plants (Bardos et al., 2020a; Song et al., 2019). Nevertheless, natural decontamination processes or bioremediation are regarded as a 'regulating ecosystem service' performed by microorganisms, earthworms and other soil organisms functioning in healthy soils; therefore, a high diversity and biological activity within soils, especially at the level of chemical engineers, but also of ecosystem engineers, is indispensable to ensure this essential service (FAO et al., 2020; Orgiazzi et al., 2016; Turbé et al., 2010).

2.4 Phytomanagement

A promising new direction in the application of GRO is **phytomanagement**; commonly defined as *"the long-term combination of profitable crop production with gentle remediation options (GRO) leading gradually to the reduction of contaminant linkages due to metal(loid) excess and restoration of ecosystem services"* (Cundy et al., 2016; GREENLAND, 2014a, 2014b; Robinson et al., 2009). Phytomanagement has been demonstrated to be an effective strategy for sustainably managing and monitoring risks posed by a wide variety of contaminants (Bardos et al., 2020a; Burges et al., 2018; Cundy et al., 2016; Gerhardt et al., 2017; Robinson et al., 2009), improving soil functions and ecosystem services (Burges et al., 2018, 2016; Cundy et al., 2016; Epelde et al., 2014b, 2009a, 2009b; Gómez-Sagasti et al., 2012; Kidd et al., 2015; Mench et al., 2010; Touceda-González et al., 2017a), and generating profits where local conversion chains are present to value biomass (Andersson-Sköld et al., 2014; Conesa et al., 2012; Cundy et al., 2016; Evangelou et al., 2012; GREENLAND, 2014a). This can either be a short-term, temporary solution (e.g. as a 'holding strategy' until a different site use is decided) or as a long-term land management strategy for a 'soft' end use as for green land uses and recreational greenspaces (Bardos et al., 2020a, 2016; Cundy et al., 2016). A requirement for successful phytomanagement, therefore, is that it should either cost less than other remediation alternatives or be a profitable operation (Conesa et al., 2012; Robinson et al., 2009). Furthermore, phytomanagement should entail the best site-specific, cost-effective management option for managing risks at a site, and can be wholly based on GRO but it does not proscribe the use of other remediation technologies to achieve the best outcome, e.g. as part of a treatment chain (Robinson et al., 2009). 'Crop-based' systems for RBLM have successfully demonstrated the benefits of vegetation-, energy crop-, or generally nature-based solutions for both managing risks at contaminated sites and providing wider value including bio-based production for bioenergy and other ecosystem services (Andersson-Sköld et al., 2014, 2013a; Bardos et al., 2020a; R. P. Bardos et al., 2011; Cundy et al., 2016; Enell et al., 2016; Gomes, 2012; GREENLAND, 2014a; Schröder et al., 2018).

Best practices to create 'windows of opportunity' for successful phytomanagement have been developed and optimised in large-scale European projects (e.g. GREENLAND and PhytoSUDOE); including by means of i) enhancing standard phytoremediation strategies with soil amendments and/or bacterial inoculates and mycorrhizal fungi, ii) creating tree plantations

based on short-rotation coppicing of woody plants such as poplar and willow, iii) using high-biomass annual or perennial herbaceous species (e.g. rapeseed, sunflower, tobacco, bioenergy grasses, maize, etc.), and iv) applying best practice agronomic techniques like crop rotations, intercropping with legumes, agroforestry, cover crops, etc. to improve phytoremediation effectiveness (Garbisu et al., 2019; Gómez-Sagasti et al., 2018; GREENLAND, 2014a, 2014b; Kidd et al., 2015; Mench et al., 2019; Moreira et al., 2021, 2019).

2.5 GRO applicability

According to OVAM (2019), plant-based GRO (phytoremediation) can be used for i) the remediation of moderate, low or (potentially) high concentrations of inorganic and organic contaminants, even if they are spread over large areas; ii) the remediation of remaining contamination after removal of source zones with conventional remediation (i.e. soil polishing, treatment chains); iii) to prevent the infiltration of contaminants into groundwater or to reduce the leaching of fertilizers and pesticides into rivers and other sensitive areas; iv) to control the spreading of diffuse, non-point source contamination (e.g., air deposition); and v) to provide an active form of controlled natural attenuation. Examples of site or project conditions which do not favour conventional remediation, but may be suitable for GRO, include (Cundy et al., 2016, 2013b; GREENLAND, 2014a):

- Budgetary and deployment constraints (e.g. large areas with diffuse contamination not causing immediate concern such as abandoned railway or road corridors)
- Biological functioning is desired post-remediation (e.g. greenspaces, parks)
- Ecosystem services are highly valued (e.g. riverbank greens, urban wilderness)
- A need to restore land and a potential to produce non-food crops (e.g. for biofuels)

Typically, these constraints describe a site where a 'soft' end use is envisaged, which are well-suited for provisioning greenspace, green infrastructure, or other similar land uses which require a functioning soil ecosystem (Bardos et al., 2016; Cundy et al., 2016; GREENLAND, 2014a; Menger et al., 2013). This kind of land use can be readily incorporated into urban design and landscape architecture either on a long-term basis as a 'self-funding land management regime' (Andersson-Sköld et al., 2013b) or as an interim 'holding strategy' at vacant sites (Cundy et al., 2016). Furthermore, connected to each plant-based GRO application, a set of landscape design strategies, or 'phytotypologies', have been created for various site-specific applications that can be adapted and integrated as part of the landscape architect's toolkit (Cundy et al., 2016; Kennen and Kirkwood, 2015; OVAM, 2019).

Case studies and project profiles successfully implementing "phyto-technologies"² or "ecological revitalisation"³ have been collected by the US EPA's Clean-Up Information (CLU-IN) portal for a variety of sites including old mining areas, foundries, manufacturing facilities, refineries, landfills, military installations and tanneries, though it is stressed that these techniques can be implemented to some degree at any site (ITRC, 2009; US EPA, 2009).

² [CLU-IN | Databases > Phytotechnology Project Profiles \(clu-in.org\)](https://clu-in.org/Databases/Phytotechnology/ProjectProfiles)

³ [CLU-IN | Databases > Ecological Revitalization Project Profiles Database \(clu-in.org\)](https://clu-in.org/Databases/EcologicalRevitalization/ProjectProfilesDatabase)

3 METHODOLOGY

This chapter provides the methodology followed to achieve the overall aim and specific research objectives.

The methodology to achieve the overall aim and specific objectives of the thesis entailed building the theoretical foundation of the research through literature review, conducting interviews with experts, developing a risk management framework for GRO and then applying it in case studies. The following sections and Figure 3-1 provide additional detail as to how these parts of the work were carried out.

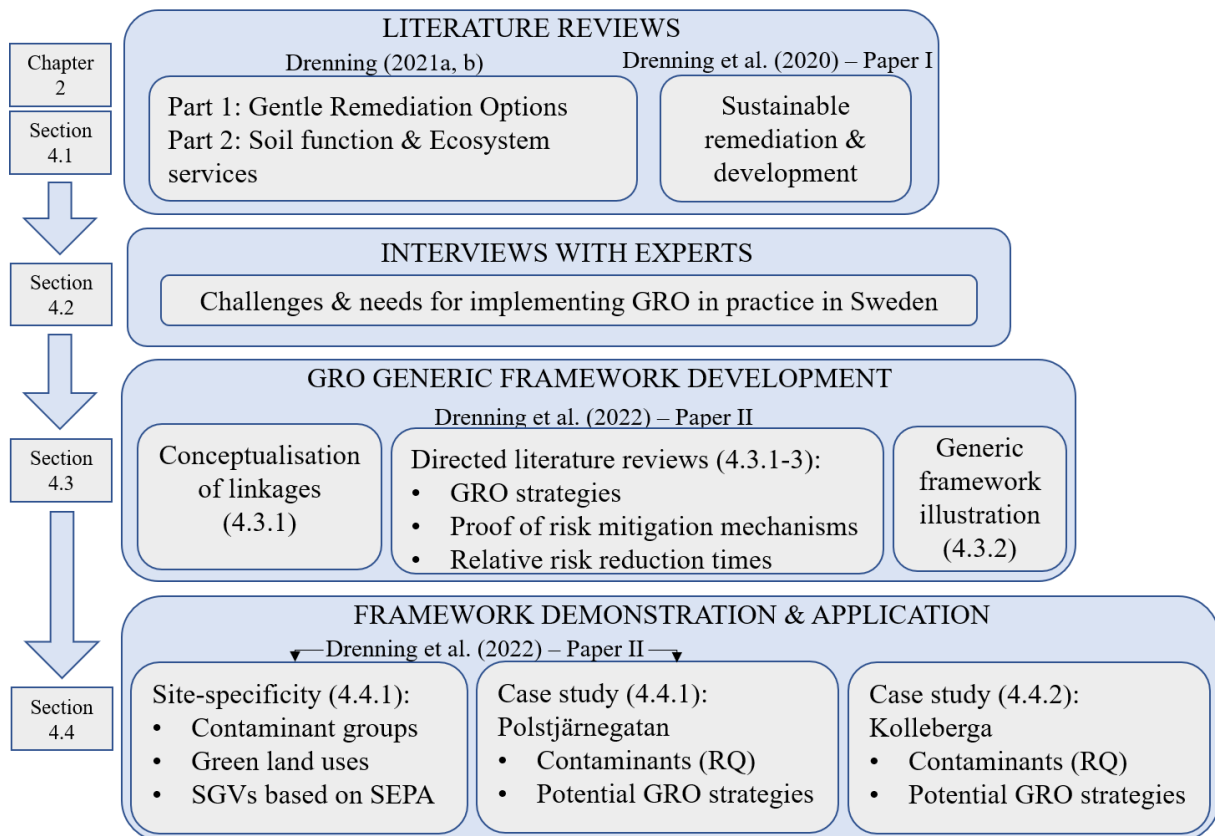


Figure 3-1. Methodology for the licentiate thesis.

3.1 Literature review

Multiple literature reviews contributed to fulfilling the overall aim and the specific objectives of this thesis. The first review in Drenning et al. (2020) (Paper I) aimed to identify key themes of intersectionality between related fields that can be used to support decision-makers by reinforcing the connections to sustainable remediation and development. Extensive, semi-systematic reviews were also carried out and targeted towards synthesising need-to-know, practical information for 1) successful and effective application of GRO (Drenning 2021a) and 2) understanding and assessing soil functions and ecosystem services primarily in the context of contaminated sites (Drenning 2021b). These reviews form the theoretical foundation of the Ph.D.-project and supported the development of the risk management framework. Further detail about the methods for reviewing literature and writing can be read in each report.

3.2 Interviews with experts

To gain a better understanding of the general state of GRO and ecosystem services, regarding awareness, potential applications at contaminated sites, etc., interviews were conducted with individual experts working as researchers, consultants, regulators and in related fields in Sweden. Interviews were not extensive and only five experts in total were interviewed, but it served to provide an understanding about challenges and opportunities in the field rather than an extensive mapping. Background information and a short questionnaire was sent to inform the respondent and for them to record their perceptions, which was then used to guide the discussion. The questions were intended to be broad and open-ended, and related to i) whether the respondent had experience working with GRO and/or ecosystem services, ii) if they believed that there was interest (growing or shrinking) concerning these concepts in their organisation or network, iii) what they saw as the major advantages or disadvantages of GRO and what roll GRO could play in contaminated site remediation, and iv) why they believed GRO were not used more often in the present-day circumstances.

3.3 Framework development

As reported in Drenning et al. (2022) (Paper II), the working process for developing the risk management framework for GRO proceeded according to the following steps:

1. Development of a conceptualisation of connections between GRO, risk mitigation mechanisms and their impact on ecological and human health risks. An extensive literature review was undertaken to identify studies that can support the hypothesised risk mitigation mechanisms. The conceptualisation is illustrated in a conceptual diagram and forms the basis for the generic framework. Mapping of the expected timeframes for effective risk reduction of different GRO and contaminant groups was based on existing literature. The time perspectives for different GRO and groups of contaminants were added to the figure which altogether forms the generic risk management framework.
2. For demonstration purposes, the following selections were made:
 - i. Based on Chowdhury et al. (2020), three green land uses were selected – Biofuel Park, Recreational Park and Allotment Gardens – which theoretically represent a "low", "medium" and "high" risk scenario, respectively. Depending on contaminants that are present, different receptors and human health exposure pathways will dominate the risk situation.
 - ii. Thirteen specific contaminants that are commonly found at urban brownfields were selected as representatives for the contaminant groups shown in Figure 2-5: 1) metal(loid)s – lead, cadmium, arsenic, copper and zinc; 2) petroleum products – PAH's (groups of light, medium and heavy density compounds) and benzene; 3) persistent organic pollutants – PCB's, dioxins and Σ DDT (including DDE and DDD); and 4) chlorinated solvents – TCE. The selection was based on that they should represent most of the different groups of contaminants (Kennen and Kirkwood, 2015; OVAM, 2019; Swartjes, 2011) and a report for the European commission on soil contamination and its impacts on human health which documents some of the top contaminants of

concern in soils (Science Communication Unit, University of the West of England, 2013).

- iii. In order to calculate land use specific SGV for the selected green land uses and contaminants, the Swedish national soil guideline value model (SEPA, 2009) was used. Relevant adjustments to the generic scenarios implemented in the model were made and based on this, the most important receptors and human health exposure pathways can be identified for each contaminant and each green land use.

3.4 Framework application

The generic framework can be adapted to account for certain site-specific considerations and envisioned future land use in order to apply the framework at any particular site. By adjusting exposure parameters used in the Swedish guideline value model (SEPA, 2016, 2009) (e.g. time on site), exposure scenarios more reflective of realistic land usage by visitors were first created for three green land uses (biofuel park, recreational park and allotment garden). Based on the SEPA model, the soil guideline values (SGV), dominating risks and corresponding exposure pathways for the three green land uses were calculated and identified for commonly detected contaminants in urban soils. Drenning et al. (2022) (Paper II) presents more information on this part of the working process and preliminary results concerning the varying risk assessment per modelled green land use exposure scenario and corresponding SGVs. The generic conceptualisation of connections between GRO, risk mitigation mechanisms and their impact on ecological and human health risks is applied to two case study sites. For either site, to account for the varying contamination levels and provide an indication of the relative risk, the Risk Quotients (RQ) for each contaminant was calculated by dividing the mean (total) concentration in the soil by either the corresponding health-based SGV or the lowest environmental SGV determined in the land-use specific SEPA model.

3.4.1 Case study site – Polsjärnegatan (Gothenburg, Sweden)

The case study site is part of a concept plan of a large-scale housing and commercial development area “Karlstaden” in Gothenburg, Sweden and site investigations have detected elevated contamination concentrations. The planned future use is a park area with new roads constructed along the edges of the site. It was initially used as a railyard for coal products and was later transformed into a golf course. The golf course closed in the early 2000s and the site has been abandoned ever since. According to the environmental investigation conducted at Polstjärnegatan, the site is characterised by several small hotspots resulting from illegal cable burning with high contamination levels, and the rest of the area with lower contamination, primarily in the upper soil layer, 0 – 0.7 m (Kaltin and Almqvist, 2016). The primary contaminants are metal(loid)s (As, Cu, Pb, and Zn), petroleum products (primarily PAHs with high molecular weight) and PCB. As the hotspots have concentrations at levels corresponding to hazardous waste according to Swedish legislation, remediation by excavation some other type of faster source removal technology at those spots is likely needed but requires a site-specific risk assessment. For the demonstration of the suggested framework, the rest of the area is considered, where contamination levels are lower. See Drenning et al. (2022) (Paper II) and

the attached Supplementary Material for additional information on the site including contaminant concentrations and RQ calculations.

3.4.2 Case study site – Kolleberga (Ljungbyhed, Sweden)

Kolleberga is a former tree nursery located in Ljungbyhed, Sweden. The site was operational since the 1950's to cultivate different pine, spruce and fir tree species for research and commercial purposes. DDT was used to manage weeds and pests across the site by both dipping tree cuttings into vats of liquid DDT and spraying broadly over the fields. DDT usage was stopped in the middle of the 1970's and today the site's operations are largely discontinued. However, due to DDTs persistence in the environment, the site is still contaminated with DDT (and traces of other pesticides) over large areas with medium to low concentrations in the topsoil. DDT concentrations in groundwater and local surface water are not elevated but there is a risk of spreading to these recipients. The main agricultural fields cover an area of approximately 18 hectares and has a mean soil contamination level of about 7.25 mg/kg total DDT (measured as a sum of DDT and its cometabolites) (Sandström et al., 2020), which is above generic guideline values of 0.1 and 1 mg/kg for sensitive and less sensitive land use respectively (SEPA, 2009), but varies across the site to a max level of 23 mg/kg. There is also a 'hot spot' (where the dipping operations were purported to occur) where significantly higher concentrations were measured (mean 35 mg/kg, max 227 mg/kg). Detailed ecological and human health risk assessments have been carried out by Tyréns (Sandström et al., 2020). Site-specific guideline values were created for the site by adjusting the generic guideline values based on prevailing site conditions which could be used to determine site-specific risks instead of those built on generic assumptions. The site-specific SGV for the environment matched the generic DDT SGV for sensitive land uses (0.1 mg/kg) but the SGV for human health was adjusted to 16 mg/kg. The results from the risk assessment determined that the human health risks are low/acceptable, but that there is an accumulation of DDT in earthworms which could potentially negatively affect them and other soil organisms thereby inhibiting soil functioning. There is also judged to be a risk of secondary poisoning higher in the food chain to shrews and other small animals for which earthworms are food, predatory birds hunting in the area and possibly grazing animals.

4 RESULTS




This chapter summarises the main results of the Ph.D.-work thus far, starting with a summary of the possibilities and main benefits of GRO in relation to sustainable remediation and development and risk management. Separate sections present results from interviews with experts, the risk management framework and supporting evidence, and a demonstration of the framework by applying on two case study sites.

4.1 Potential applications of GRO

4.1.1 GRO strategies for sustainable remediation and development

As discussed in Drenning et al. (2020) (Paper I), brownfield sites represent important land and soil resources and provide significant opportunities in urban, peri-urban and even rural areas to meet national and international environmental goals. GRO as innovative remediation techniques and alternative land management strategies offer many direct and co-benefits in relation to sustainable remediation and development, some of which are summarised in Table 4-1.

Table 4-1. GRO strategies for sustainable remediation and development – summary of benefits. Icons are shown for SDGs identified in Drenning et al. (2020) and additional related SDGs are listed below these.

	Environmental	Economic	Social
GRO benefits	<ul style="list-style-type: none"> • Low-impact in-situ remediation of many contaminants • Mitigated environmental impacts of remediation and urban land use • Preserving/improving soil and land resources • Elements of green infrastructure • Provide/restore/maintain ecosystem services and soil function • Nature-based solutions • Resilience to climate change impacts and natural disasters • Carbon sequestration 	<ul style="list-style-type: none"> • Significant cost savings (ca. 50%) compared to conventional remediation alternatives • Enables remediation of low- and moderate-risk contaminated sites at lower cost • Highly useful for sites of low land value or where conventional remediation techniques are unsuitable • Useful as part of a treatment train • Potential for profitable biomass production (e.g. bioenergy) 	<ul style="list-style-type: none"> • Improved urban liveability • Allow sites to be used as formal greenspace and other 'soft' uses • Vegetated brownfields also provide informal recreational space and other citizen benefits • Aesthetically pleasing remediation • Potential part of landscape architecture toolkit
Related SDGs	<div> <div>  <p>11 SUSTAINABLE CITIES AND COMMUNITIES</p> </div> <div>  <p>13 CLIMATE ACTION</p> </div> <div>  <p>15 LIFE ON LAND</p> </div> </div> <p>poverty elimination (1), zero hunger (2), good health and well-being (3), protecting children and women (5), supplying safe drinking water (6), increasing security and resilience of cities (11), mitigating climate change (13), preventing loss of aquatic biodiversity (14), preventing land degradation and loss of terrestrial biodiversity (15)</p>		
Relevant political goals & EU directives	European Green New Deal, Biodiversity Plan to 2030, Soil Thematic Strategy, Circular Economy, Zero Pollution Action Plan		

4.1.2 GRO applications for situational risk management

A summary table compiling the main situational applications for GRO, corresponding GRO mechanisms, targeted contaminants, typical plants used, and the general strategy and objectives

are listed in Table 4-3. For each application, the 'main' or 'secondary' GRO mechanisms vary depending on the site-specific purpose and contaminants present at a site and whether the remediation objective is intended for source removal, stabilisation, containment, as part of a treatment chain or complementary soil polishing, etc. Bioremediation, mycoremediation and vermiremediation not included in this table as they do not fit neatly into the same application classification; however, they are also viable to implement in a variety of situations to manage a wide range of contaminants. Bioremediation, for instance, can be used to degrade various organic contaminants in soil and is especially well-suited for groundwater and surface water whereas myco- and vermiremediation are better suited for soil contamination.

4.2 Challenges and possibilities for GRO in Sweden: Interviews with experts

Interviews were conducted with a small, non-extensive group of experts (5) whose responses have been anonymised and summarised (Table 4-2). In general, the responses can be separated into whether they address possibilities or challenges which then have been grouped according to four main aspects: general, practical, knowledge and development.

Table 4-2. Summary table of compiled and anonymised answers from interviews with experts separated into four main aspects: General, Practical, Knowledge and Development with a Summary question. Bold text indicates a frequently stated or important implication.

Aspects	Possibilities	Challenges
General	<ul style="list-style-type: none"> • Holistic view → resilience, multi-function • Focus on socio-economic value • Considerable interest – but time is an important question • Risk 'reduction' vs 'elimination' → reduce resource intensive remediation 	<ul style="list-style-type: none"> • Time and uncertainties → limitations • No clear recipient/actor in society who is responsible for long-term knowledge development (or for the entire soil ecosystem) and for using the results • Business economic risk → low or non-existent incentives for businesses • Lack of knowledge
Practical	<ul style="list-style-type: none"> • Demands practical experience and long-term monitoring/follow-up • Bad (in the branch) at showing how ES are connected to contaminated sites but the interest exists • GRO are appropriate at sites with low risk and no immediate development or time pressure 	<ul style="list-style-type: none"> • Must find a fitting ES typology and valuation methods • Perceived limited applications • Long-term effects/risks → who has responsibility in 10, 20+ years? What happens in the future? • Landowners need/want to manage risks (often immediately)
Knowledge	<ul style="list-style-type: none"> • Bioavailability and a 'risk index' (not just a number) → a measurement must be fast, easy and easily understandable • Technological development → niche technique 	<ul style="list-style-type: none"> • Lack of knowledge regarding both ES and GRO (especially within contaminated sites context) • Uncertainties → how effective are GRO? Must show that they work • Difficult to measure what is possible (results) → demonstrate effectiveness
Development	<ul style="list-style-type: none"> • Transition from pure remediation focus → soil improvement, e.g. 'what is left after remediation?' → upgrading degraded land • New trend to focus on soil as a valuable resource • SEPA evaluation of remediation guidance documentation concluded that 'ES must permeate remediation work' 	<ul style="list-style-type: none"> • 'Status quo bias' → resistance to change and overreliance on conventional methods • Soil as a 'property' not an 'asset' → soil quality usually not accounted for (mostly just talk) • Soil improvement is an entirely different world than remediation

In general, the respondents indicated that while there may not be much current implementation of GRO or ES assessment at contaminated sites in Sweden, they note a growing interest in

including these in work with contaminated sites and are convinced that the field is moving that direction (Table 4-2). All respondents were positive concerning trends towards GRO, ES, and NBS and believe that the timing is right for uptake and application of these concepts. Respondents that work with regulatory and governmental agencies noted that there is a growing interest at the EU level (but more slowly in Sweden) to shift focus from a focus on simply decontamination to emphasise instead risk-reduction and placing greater value on "what is left" after remediation. They referred to the new emphasis to "upgrade degraded land" more broadly and account for improving soil health and function. However, another respondent stated that "soil improvement is an entirely different world than remediation", which could indicate that wider, paradigm shifting ambitions may not be quickly adopted by experienced remediation practitioners. The same respondent issued a word of caution and clarification to state that "what drives contaminated site management is the need to manage risks, which must be answered directly and shown definitively." This sentiment was reflected in other respondents who all mentioned that the uncertainties and lack of knowledge surrounding GRO is a significant obstacle to widespread adoption. Furthermore, in Sweden, there is not clear societal recipient who has responsibility for the long-term development of knowledge or for using the results of research projects, and many governmental agencies may feel that this lies outside of their purview.

Table 4-3. GRO applications for situational risk management, summarised and modified from (ITRC, 2009; Kennen and Kirkwood, 2015; OVAM, 2019; Schnoor, 1997).

Application	GRO mechanism	Media	Contaminants	Example sites	Typical plants	Strategy/Objectives
Vegetation covers (in combination with soil amendments) for stabilisation of soil, sediments and infiltration control	Main: Phytostabilisation, Immobilisation/Stabilisation Secondary: Phytodegradation, Rhizodegradation, Phytovolatilisation, Phytoextraction, Phytohydraulics	Soil Sediment Groundwater Surface water (water vector)	Valid for most (if not all) contaminants but most relevant for non-bioavailable contaminants. Inorganic: Metal(loids) (e.g. Pb), salts, nutrients (P), radionuclides Organic: persistent organic pollutants (POPs) and other hydrophobic organics (e.g. PCBs, PAHs, dioxins, furans, PCP, DDT, dieldrin)	Landfills (to cover and control leachate), former mining sites and tailings, petroleum extraction sites and refineries, military bases and firing ranges, large agricultural fields, railway and roadway corridors, underutilised industrial areas and other marginalised lands	<ul style="list-style-type: none"> Thick, densely planted, densely fibrous and deep-rooted herbaceous (ideally perennial) vegetation cover (e.g. fescue, ryegrass, bent grass mixes) Excluding (non- or low-accumulating) plant species Leguminous species (e.g. clover, alfalfa) Plant species with high evapotranspiration rates 	<p>Hold contaminants on site and reduce water infiltration and leaching via:</p> <ul style="list-style-type: none"> Reducing bioavailability/solubility of contaminants. Combining with soil amendments to improve effects. Revegetation of barren sites – <i>provide ecosystem services</i> <p>Vegetation cover also provides risk reduction by erosion control, hydraulic control, dust control, and managing receptor access to contaminants.</p>
Vegetation covers for phytoremediation (extraction, degradation or volatilisation)	Main: Phytoextraction, Phytodegradation, Rhizodegradation, Phytovolatilisation, Phytostabilisation Secondary: Phytohydraulics	Soil Sediment Groundwater Surface water (water vector)	Inorganic: Readily extractable metal(loids) such as As, Cd, Co, Cu, Ni, Se, Zn; radionuclides (Ba, Ce, Cs, Sr, U), nutrients (N and P) Organic: pesticides/herbicides (e.g. atrazine, alachlor, dieldrin); petroleum products such as BTEX, MTBE, aliphatics and PAHs; chlorinated solvents (e.g. TCE, PCE, VC and other VOCs); explosives (TNT, RDX); POPs (DDT, PCB)	Large areas or residences with As, Cd, Ni, Se, Zn or other readily extractable metal(loids), long-term agricultural field remediation, perimeters of gas stations, auto-repair shops, dry cleaners, community gardens, military bases and firing ranges, former industrial sites/salvage yards, large industrial areas of metal smelters, railway and roadway corridors, underutilised industrial areas and other marginalised lands	<p>Planting mix that can be a <i>multi-mechanism design</i> to degrade organics and extract bioavailable inorganics:</p> <ul style="list-style-type: none"> Hyperaccumulators (of As, Cd, Ni, Se, Zn) Metal accumulating and high biomass producing trees (e.g. willow, poplar) and field crops/grasses Tree/shrub species for degrading organics Leguminous plants and grasses Phenolic releasing trees (mulberry, apple) 	<p>Gradual source removal via:</p> <ul style="list-style-type: none"> Bioavailable contaminant stripping of inorganic contaminants Enhanced biodegradation/volatilisation of organic contaminants – improved by biostimulation bioaugmentation and selecting plants for root exudates and associated microbes Revegetation of barren sites – <i>provide ecosystem services</i> <p>Vegetation cover also provides risk reduction by erosion control, hydraulic control, dust control, and managing receptor access to contaminants.</p>
Phytoremediation tree stand	Main: Phytohydraulics, Phytodegradation, Rhizodegradation, Phytovolatilisation Secondary: Phytoextraction, Phytostabilisation	Soil Sediment Groundwater Wastewater	Inorganic: Metal(loids) (Cu, Cd, Pb), salts Organic: dissolved organic compounds including petroleum products (e.g. BTEX, MTBE, aliphates, and petroleum hydrocarbons including gasoline-, diesel- and oil-range organics)	Gas stations, auto-repair shops, dry cleaners, urban industrial site perimeters, funeral homes and cemetery buffers, agricultural hedgerows, leaking underground storage tanks, fertilizer spills	<p>Phreatophyte and deep-rooting or tap rooting trees, shrub species with high evapotranspiration rates (e.g. willow, poplar, aspen).</p> <p>Certain species are better suited for degradation depending on root exudates and associated microbes.</p>	<ul style="list-style-type: none"> Lateral migration control of contaminants spreading in groundwater. Targeted remediation of contaminants in groundwater and deeper soil layers with deep-rooting trees – improved by biostimulation, bioaugmentation and selecting plants

			and chlorinated solvents (e.g. TCE, PCE, DCE, VC)			for root exudates and associated microbes for specific contaminants.
Hydraulic barrier	Main: Phytohydraulics Secondary: Phytoextraction, Phytodegradation, Rhizodegradation, Phytostabilisation, Phytovolatilisation	Groundwater	<i>Highly applicable for contaminated groundwater plumes</i>	Rail, military and industrial facilities or dry cleaners with chlorinated solvent and VOC plumes (e.g. TCE, PCE); leaking underground storage tanks, gas stations and petroleum refineries with BTEX or MTBE plumes		<ul style="list-style-type: none"> Primarily intended for controlling the direction and velocity (flux) of the groundwater to hydraulically control a contaminated groundwater plume.
Riparian buffers and other buffer zones (e.g. motorways and fields)	Main: Phytohydraulics, Rhizofiltration Secondary: Phytoextraction, Phytostabilisation, Phytodegradation, Rhizodegradation, Phytovolatilisation	Soil Sediment Groundwater Surface water (water vector)	Inorganic: Nutrients (N and P) from fertiliser and metals from agricultural and road runoff Organic: pesticides, TCE, PCE, BTEX, MTBE, DRO	Riparian buffers by water bodies, corridor buffers for roadsides, railroads, industrial areas and agricultural plots, site perimeter buffers	Diverse, mixed planting of trees, shrubs, herbaceous, coastal and aquatic plants in the various 'riparian planting zones'	<ul style="list-style-type: none"> Erosion control and hydraulic control of shallow groundwater to prevent contaminants spreading into nearby water bodies through contaminated groundwater plumes, runoff and erosion.
Stormwater biofilters (rain garden, bioswale, green roof)	Main: Rhizofiltration, Phytohydraulics Secondary: Phytostabilisation, Phytodegradation, Rhizodegradation, Phytovolatilisation	Sediment Surface water Stormwater (water vector)	Inorganic: Nutrients (N and P) from fertiliser and metals from agricultural and road runoff; Organic: pesticides, TCE, PCE, BTEX, MTBE, DRO	Roadsides and parking lots, agricultural fields, construction areas, golf courses, convention centres, commercial and industrial buildings and infrastructure	Aquatic macrophytes (bullrush, cattail, coontail, pondweed, arrowroot, duckweed, common reed, yellow iris); turf and ornamental grasses; perennial flowers, shrubs and trees	<ul style="list-style-type: none"> Erosion and hydraulic control of shallow groundwater and prevention of contaminants spreading into nearby water bodies in runoff and erosion. Manage stormwater volume close to site and prevent contaminant mobilisation.
Constructed treatment wetland/aquatic plant lagoon	Main: Rhizofiltration Secondary: Phytostabilisation, Phytodegradation, Rhizodegradation, Phytovolatilisation	Sediment Surface water Stormwater Wastewater Irrigation water	Inorganic: continuous removal of metal (loid) and nutrient solutes in water (e.g. As, Cd, Cu, Cr, F, N, P, Pb, Se, Zn), cyanide, radionuclides Organic: hydrophobic organics such as PAHs, PCBs, dioxins, furans, PCP, DDT, dieldrin, explosives	Wastewater treatment, golf courses, contaminated groundwater plumes, landfill leachate, stormwater wetlands	Aquatic macrophytes: Emergents (bullrush, cattail, coontail, pondweed, arrowroot, duckweed, common reed, yellow iris, water hyacinth) and Submergents (algae, stonewort, parrot feather, Eurasian water milfoil, hydrilla)	<ul style="list-style-type: none"> Clean and filter contaminants from surface water, stormwater and wastewater Prevent soil erosion by water runoff and protect other water bodies by limiting contaminant spreading and reducing concentrations in effluent

4.3 A risk management framework for gentle remediation options (GRO)

As reported in Drenning et al. (2022) (Paper II), a risk management and communication framework was developed to clarify the connections between GRO, risk mitigation mechanisms and their impact on ecological and human health risks. The results are reported here in brief for three parts: 1) the conceptual diagram to illustrate the connections, 2) the literature review to identify GRO risk mitigation mechanisms, compile studies that support them, and map effective risk reduction times for each GRO strategy and group of contaminants; and 3) the resulting generic risk management framework for GRO.

4.3.1 A conceptualisation of GRO connections for risk management

Where there are verifiable contaminant S-P-R linkages, thus posing exposure risks to a receptor, GRO can be used to manage the risks by breaking these linkages (Figure 4-1). In summary, three primary 'risk mitigation mechanisms' can be attributed to GRO:

1) *Bioavailability and solubility reduction* – mitigating the risks posed to humans by stabilising or immobilising soil contaminants through physical and/or bio(chemical) results in reduced bioavailability and mobility thereby mitigating exposure risks to humans. (Friesl-Hanl et al., 2017; GREENLAND, 2014a; Mench et al., 2010; OVAM, 2019). Risks to the environment could also be mitigated by this mechanism through reducing the readily available concentration of contaminants in soil pore water thus limiting mobility (e.g. leaching to groundwater) and exposure to ecological receptors in soils and local surface waters (GREENLAND, 2014a; Mench et al., 2010; Quintela-Sabarís et al., 2017; Touceda-González et al., 2017b).

2) *Source removal – plant uptake, degradation, volatilisation* – removal or degradation of the bioavailable pool of inorganic and organic contaminants greatly mitigates (or altogether eliminates) the risks posed to humans and the environment (Cundy et al., 2016; GREENLAND, 2014a).

3) *Secondary effects by vegetation cover* – revegetation at a contaminated site, usually combined with application of soil amendments, is often a remediation strategy in and of itself and is a central feature of ecological restoration and phytostabilisation strategies. Vegetation cover can also provide risk management via dust control, erosion control, hydraulic control and managing receptor access to the subsurface (Cundy et al., 2016; Epelde et al., 2009b; Gerhardt et al., 2017; GREENLAND, 2014a, 2014b; Mench et al., 2010; Monica O Mendez and Maier, 2008; OVAM, 2019; Vangronsveld et al., 2009; Wong, 2003).

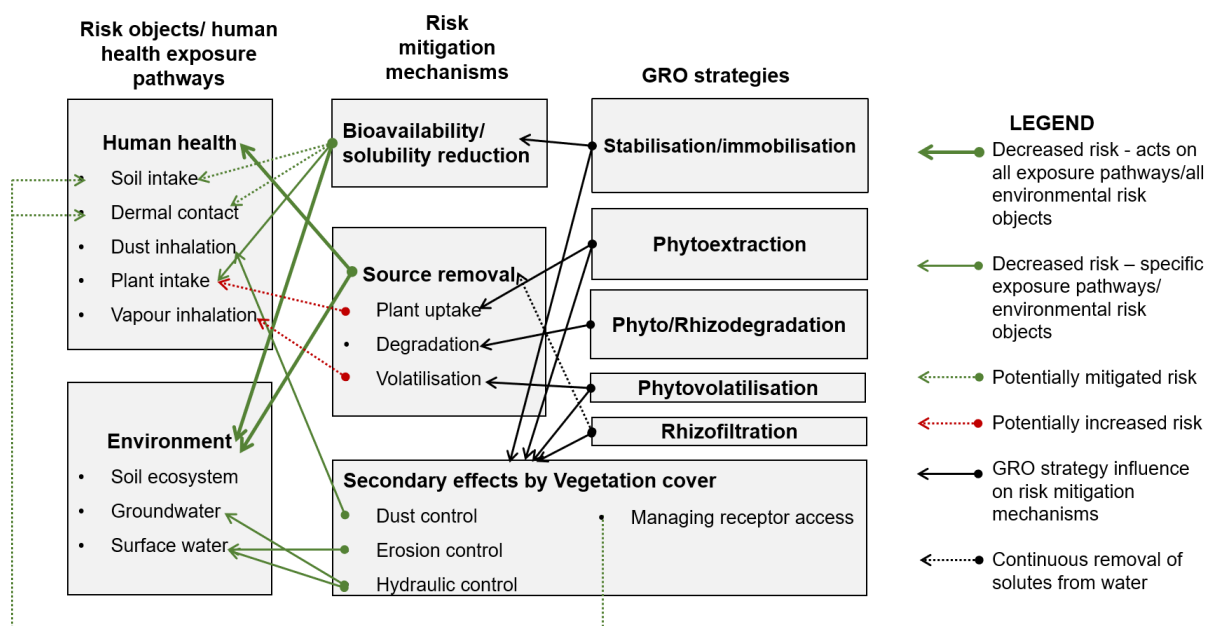


Figure 4-1. Conceptualisation of the connections between GRO strategies, corresponding risk mitigation mechanisms and effects on risk objects and human health exposure pathways.

Each of the above GRO mechanisms contribute to mitigating risks at contaminated sites by acting on specific exposure pathways to protect risk objects such as human health and sensitive environmental receptors (Table 4-4), which is further described in Table 2 in Drenning et al. (2022) (Paper II).

Table 4-4. Abbreviated table depicting connections between GRO risk mitigation mechanisms, exposure pathways and risk objects. Risk mitigation mechanisms are separated into categories corresponding to the black arrows shown in Figure 4-2. Red text indicates that the risk mitigation mechanisms potentially create an increased risk along a certain pathway. *Italic text indicates that no evidence was found in literature. Modified from (Drenning et al., 2022).*

Risk object	Exposure pathway	Description of risk mitigation mechanisms with supporting studies
<i>Risk mitigation mechanism: bioavailability and solubility reduction</i>		
Human health	Soil intake	Reducing bioavailability/solubility of contaminants using soil amendments and/or plants to immobilise in soil or plant roots, and potentially decreasing oral bioaccessibility of e.g. metals using certain soil amendments and plants though the effectiveness of this is still uncertain and requires further study (Foucault et al., 2013; Friesl-Hanl et al., 2017; Gray et al., 2006; GREENLAND, 2014a; Kumpiene et al., 2019; Mench et al., 2010, 2006; OVAM, 2019; Paltseva et al., 2020; Pelfrène et al., 2015; Sanderson et al., 2015).
	Dermal contact	Reducing bioavailability/ solubility of contaminants could potentially reduce the uptake of contaminants by absorption through the skin – <i>no studies were found to support this mechanism.</i>
	Plant intake	Reduces uptake into plants by lowering soluble, phytoavailable fraction of contaminants through the use of amendments and/or non- (i.e. excluding) or low-accumulating plant species (Ciadamidaro et al., 2019; Enell et al., 2016; Friesl-Hanl et al., 2017; GREENLAND, 2014a, 2014b; Kidd et al., 2015; Mench et al., 2010; OVAM, 2019; Tang et al., 2012; Vangronsveld et al., 2009)
Environment	-	Mitigating risks to environmental receptors by reducing the readily available of concentration of contaminants in soil pore water thus limiting mobility (e.g. leaching to groundwater) and exposure to sensitive receptors in soils and local surface waters (Andersson-Sköld et al., 2014; Enell et al., 2016; GREENLAND, 2014a; Mench et al., 2010; Quintela-Sabaris et al., 2017; Touceda-González et al., 2017a). Also, reducing toxic pressure can have a demonstrable positive effect on soil quality as measured by microbial indicators and ecotoxicity tests (Burges et al., 2017, 2016; Denyes et al., 2016, 2013; Epelde et al., 2014b, 2008; Gómez-Sagasti et al., 2012; GREENLAND, 2014b, 2014a; Kidd et al., 2015; Kumpiene et al., 2009; Quintela-Sabaris et al., 2017; Touceda-González et al., 2017b, 2017a)
<i>Risk mitigation mechanism: Source removal – plant uptake, degradation, volatilisation</i>		
Human health and Environment	-	Removal or degradation of the bioavailable pool of inorganic and organic contaminants greatly mitigates (or altogether eliminates) the risks posed to humans and the environment (Cundy et al., 2016; GREENLAND, 2014a, 2014b).
Human health	Plant uptake	Potentially introducing risks to humans or biological receptors (e.g. grazing animals) by increasing contaminant concentrations in plants or creating an 'attractive nuisance' where contaminants are more readily available than if the site were capped (Cundy et al., 2016; GREENLAND, 2014b, 2014a; Wagner et al., 2016). However, the potential added risk can in-turn be avoided by e.g. changing land use from producing food crops to bioenergy crops, pre-cultivating or co-cropping accumulating species with non-accumulating or excluding species (i.e. phytoexclusion) (GREENLAND, 2014b; Greger and Landberg, 2015; Kidd et al., 2015; Tang et al., 2012)
	Volatilisation	Could exacerbate the risks posed by vapour inhalation at a site if this is a dominant risk pathway, dependent upon the contaminant's volatility (GREENLAND, 2014a; Mench et al., 2010; OVAM, 2019; Vangronsveld et al., 2009). However, it is possible to bioaugment the plant microbiome to inoculate with bacteria that are capable of complete degradation of VOCs (OVAM, 2019; Weyens et al., 2009b, 2009a).
<i>Risk mitigation mechanism: Secondary effects by vegetation cover</i>		
Human health	Soil intake, dermal contact	Managing receptor access to the subsurface
	Dust inhalation	Dust control – reducing mobilisation by wind erosion to decrease total dust flux and fine particulates (PM ₁ , 2.5, 4)
Environment	Surface water, groundwater	Hydraulic control – using deep-rooted plant species to influence the direction and flow of groundwater and reducing contaminant flux via leaching or lateral spreading via groundwater to nearby water bodies
	Surface water	Erosion control – reduced horizontal migration of contaminants due to erosion from stormwater runoff and other physical processes

No studies were found to support whether a reduction bioavailability/solubility can mitigate exposure via dermal contact, so it is therefore regarded as a 'potentially mitigated risk' in the framework and specified with dotted green arrows in Figure 4-2. However, as indicated, this exposure pathway (and soil intake) can be effectively mitigated by having dense vegetation cover, or other barriers, to prevent contact with soil. Source removal by plant uptake or volatilisation could potentially increase the exposure risks to humans in certain situations; therefore, these have been marked with red text in Table 4-4 and red arrows denoting 'potentially increased risk' in Figure 4-2.

4.3.2 Approximating the timeframe for GRO strategies

The 'relative risk reduction time' for each GRO strategy has been estimated, based on (Kennen and Kirkwood, 2015; OVAM, 2019), and added to the generic framework (the colours and time categories correspond with Figure 2-5). Time is separated into three broad ranges: 1) *less time* (1 – 10 years), 2) *more time* (10+ years), and 3) *can potentially take decades*. In Figure 2-5, the 'relative remediation time' is estimated based on the approximate time for full source removal (e.g. via extraction or degradation) and does not provide an estimation for other risk reduction strategies such as stabilisation and vegetation cover. To address this limitation, the relative time perspectives in Figure 2-5 are expanded to also include complementary risk reduction strategies (i.e. stabilisation/immobilisation, rhizofiltration and vegetation cover). The relative risk reduction time for these strategies has been estimated to be mostly similar because the time required for the onset of risk mitigation is dependent on the time it would take for vegetation to establish or for amendments to alter soil properties. Based on literature review, vegetation establishment can be separated into three time ranges depending on plant species (shown in Figure 4-2 and discussed here as different shades of colour):

1. Quick (lightest shade) – soil amendments and fast-growing species like grasses, herbaceous species and annuals crops can provide risk mitigation within 6-8 weeks.

Medium – shrubs take longer to establish and can provide wider, more lasting risk mitigation within 1-2 years.

Slower (darkest shade) – trees provide the most extensive risk mitigation with roots able to reach down to deeper soil layers but even fast-growing tree species like willow and poplar can take from 2-4 years to establish.

For the quickest risk mitigation, soil amendments (e.g. biochar) used separately or in combination with fast-growing grasses can provide relatively 'instant' effect. For example, biochar has been demonstrated to reduce the bioavailability of PCB and DDT, thus having an ameliorating effect on earthworms, within 50 days (Denyes et al., 2016, 2013). Also, rhizomatous grasses have been recommended to quickly provide soil cover and limit the dispersal of soil particles whilst shrubs and trees establish (Mench et al., 2010; OVAM, 2019). Other fast-growing crop species such as tobacco, sunflower, mustard, willows and poplars can also provide rapid risk mitigation and typically produce high quantities of biomass, which is advantageous for phytoextraction (Herzig et al., 2014; Mench et al., 2010, 2018; OVAM, 2019; Thijs et al., 2018). For stabilisation purposes, it has been estimated that phytostabilisation of

metal(loid)s using perennial tree species like willow and poplar can generally take 2-4 years but can vary between contaminant and plant species (Robinson et al., 2006). Rhizofiltration risk mitigation is also dependent upon vegetation establishment though it varies in application (e.g. as constructed wetlands, wastewater irrigation or runoff filters), and has been demonstrated to reduce contaminant concentrations in water outflow within 1-2 years as part of an 'integrated phytomanagement system' (ANL, 2008; Cundy et al., 2020), provide ongoing treatment using willow short-rotation coppice (Dimitriou and Aronsson, 2005) and provide effective, continuous wastewater treatment (Kennen and Kirkwood, 2015; Marchand et al., 2010; Pivetz, 2001).

Adaptive GRO management is needed for all GRO strategies during their implementation, and includes long-term monitoring, watering, etc. for upkeep and to ensure the risk reduction is maintained over time. For source removal GRO strategies, adaptive management is only required during their operation until the source is removed. However, for GRO strategies that reduce risks by e.g. stabilisation/immobilisation, vegetation cover and rhizofiltration, it is important to continuously maintain and monitor the GRO while the risk mitigation mechanism is still needed to sustain the effect.

The generic risk management framework for GRO was developed to broadly conceptualise the connections between various GRO strategies, risk mitigation mechanisms, and risk objects or human health exposure pathways (Figure 4-2). Additionally, the approximate time perspectives for different GRO and groups of contaminants were added to the figure which altogether forms the generic risk management framework.

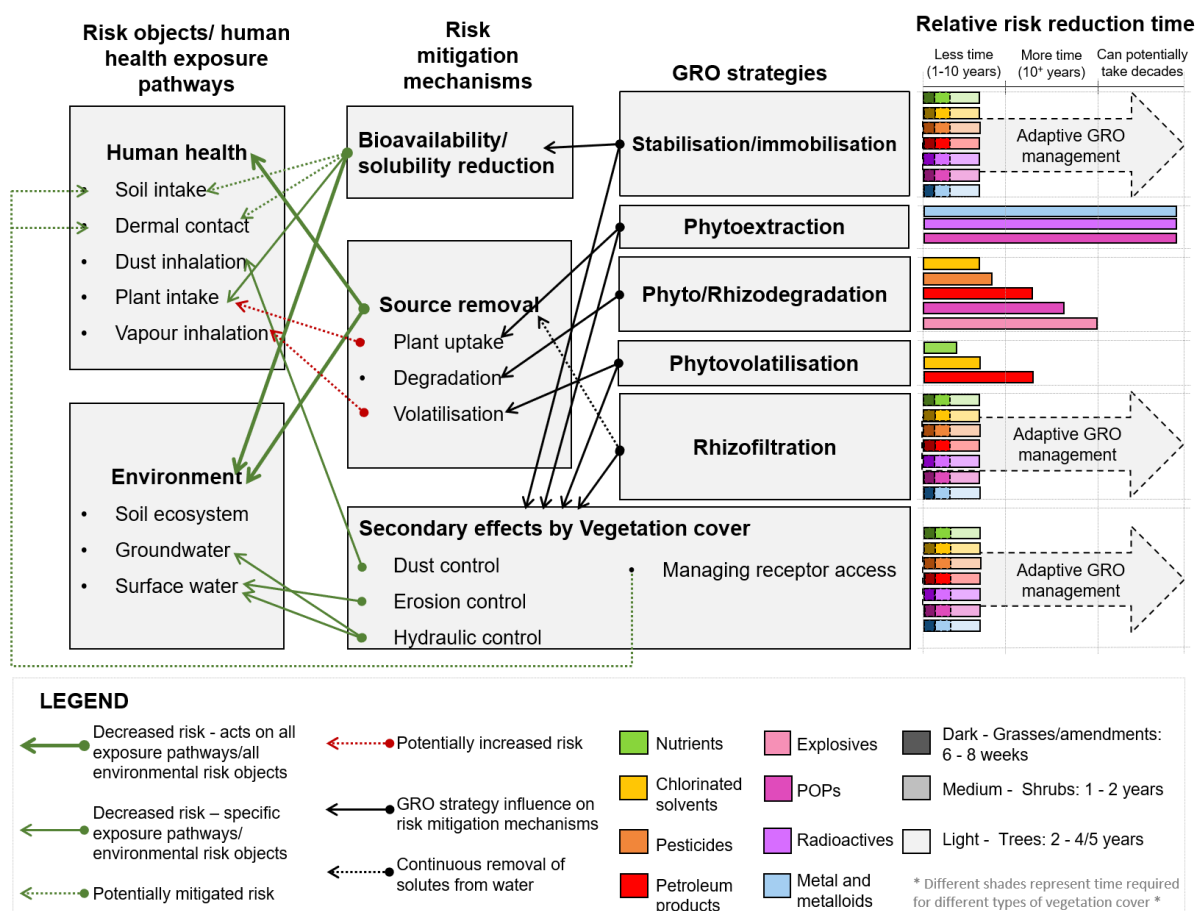


Figure 4-2. The generic risk management and communication framework for GRO with columns for Risk objects, Risk mitigation mechanisms, GRO strategies and a bar chart depicting relative risk reduction time for each GRO strategy. Relative risk reduction times are based on those shown in Figure 2-5. Relative times for stabilisation/immobilisation, rhizofiltration and vegetation cover are based on literature. Adaptive GRO management is needed for all GRO strategies during their implementation, and includes long-term monitoring, watering, etc. for upkeep and to ensure the risk reduction is maintained over time. From (Drenning et al., 2022).

4.4 Framework demonstration

Two case studies are used to demonstrate the risk management framework application for testing and reflecting upon its utility as a risk communication tool concerning GRO and identifying relevant strategies, given an envisioned land use at a particular site. Drenning et al. (2022) (Paper II) and the attached Supplementary Material presents more information on this part of the working process and results concerning the varying risk assessment per modelled green land use exposure scenario and corresponding SGVs.

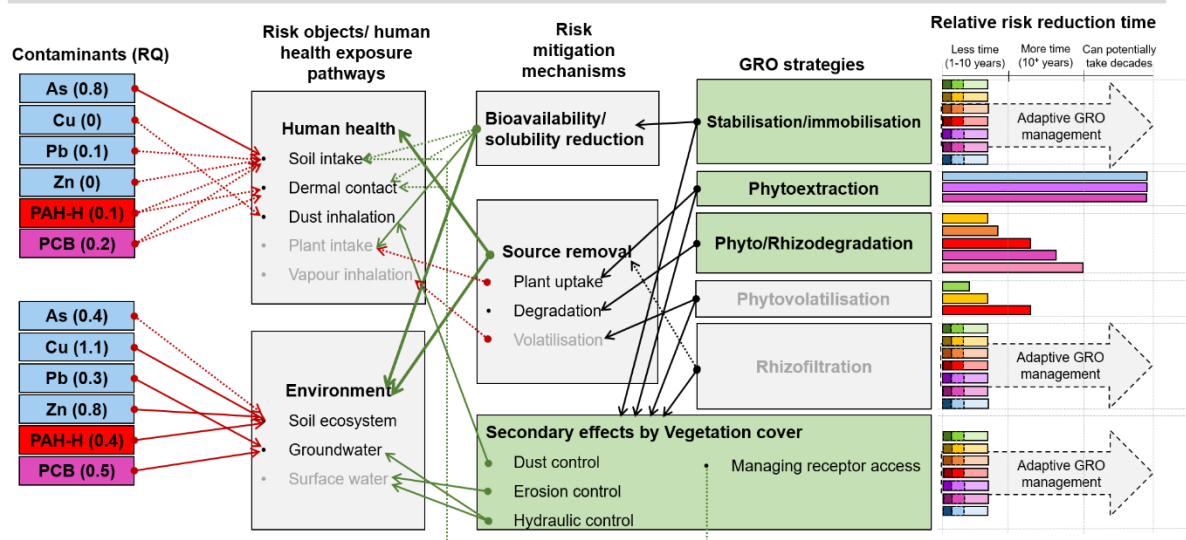
4.4.1 Polstjärnegatan

The generic framework (Figure 4-2) was adapted to include the contaminants at Polstjärnegatan and create a site-specific application of the framework for two different land uses 1) Biofuel

Park and 2) Allotment Garden (with permanent residence on site), see Figure 4-3. The dominating human health exposure pathway(s), or most sensitive environmental receptor(s), per contaminant and land use are indicated, linked with the corresponding risk mitigation mechanisms and potential GRO strategies. The calculated risk quotients are shown in the figure, and a $RQ > 1$ indicates an elevated risk (i.e. above the SGV). In Figure 4-3, for the Biofuel Park scenario, only Cu indicates a potential risk ($RQ = 1.1$, primary receptor: soil ecosystem). For the Allotment Garden scenario, the same is valid for Cu and in addition, RQs for As and PCB indicate potential human health risks (4.7 and 2.8 for soil intake and plant intake, respectively). The GRO strategies that are identified to be able to mitigate the dominating exposure pathways are highlighted in green boxes in Figure 4-3.

For the Biofuel Park, the risks posed to the soil ecosystem are of primary concern, which can be mitigated by 1) reducing the bioavailability and consequent exposure for soil organisms, and 2) removing the source of the contamination by extraction for metals or degradation for organics. A combination of these strategies could reduce the risks in the short-term (stabilisation/immobilisation) and/or achieve source removal in the longer term (extraction, degradation). The application of a 'treatment chain' could be suitable for this site entailing, for example, excavation, or some other technique to manage the source, of the highly contaminated hotspots for treatment off-site followed by use of GRO for 'soil polishing' via phytoextraction of bioavailable metal(loid)s as a risk mitigation strategy; whereby, the slightly elevated contaminant concentrations could be reduced to acceptable levels (Dickinson et al., 2009). Implementing the Biofuel Park option could potentially lead to a phytomanagement strategy that over time can allow for alternative, more sensitive land uses for unrestricted use.

Polstjärnegatan – Biofuel park



Polstjärnegatan – Allotment gardens

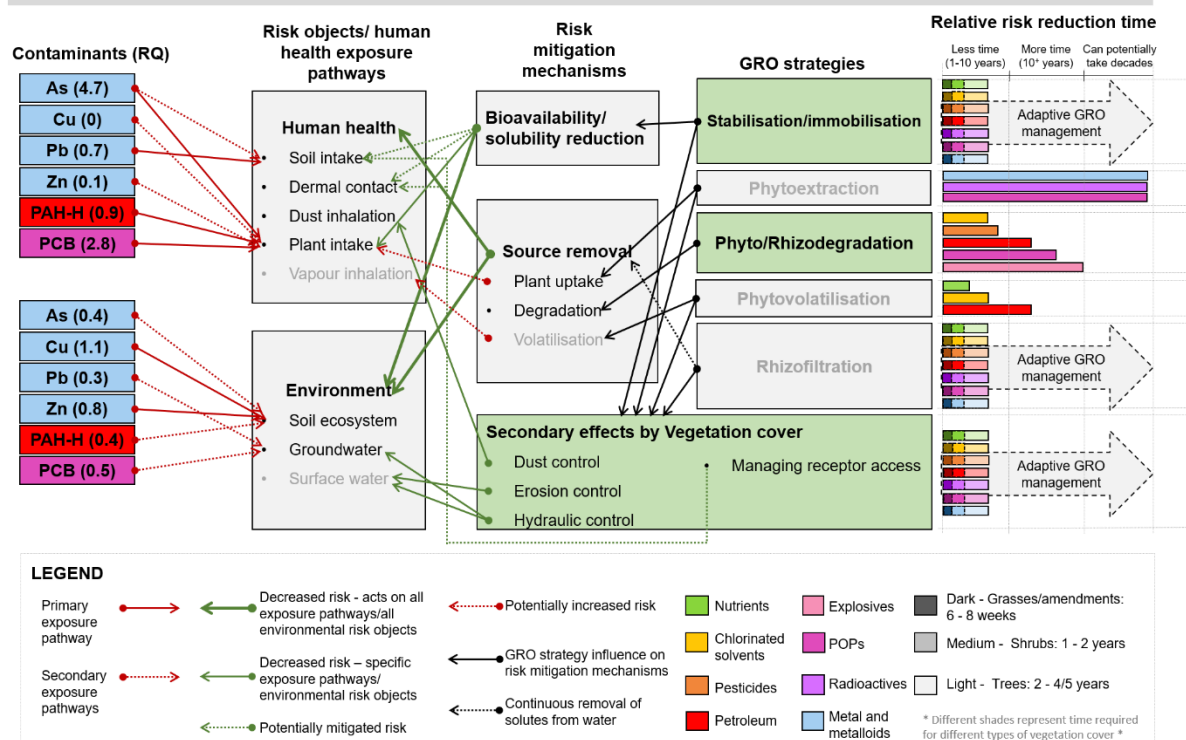


Figure 4-3. Site-specific application of the GRO risk management framework for two green land uses: Biofuel park and Allotment gardens. The contaminants detected at the site, Polstjärnegatan, and risk quotients (RQ) are included in the furthest left column and are separated into exposure pathways for human health (above) or for the environment (below).

In the Allotment Gardens scenario, As and PCB mean concentrations exceed the SGV for human health, and PAH-H is close to the threshold (RQ = 0.9). According to the SEPA model, the exposure pathway of plant intake (As, PCB & PAH-H) is of primary concern, and to a lesser extent soil intake (As). Plant intake can be mitigated by 1) reducing the bioavailability of

contaminants using amendments and plants and 2) selectively designing the vegetation cover with excluding or non-accumulating species for relevant contaminants. Soil intake can also be managed with vegetation cover consisting of dense grass species and amendments functioning as a barrier to manage receptor access to the soil and prevent humans from inadvertently ingesting the soil. Some studies indicate that GRO can also potentially reduce As oral bioaccessibility using amendments or plants, though this strategy would require a more extensive human health risk assessment and feasibility studies to confirm the effectiveness and viability as a legitimate risk reduction measure. An unrestricted Allotment Garden land use may thus not be immediately feasible and the time perspective for using GRO to meet the required risk reduction (e.g. by reducing contaminant levels via phytoextraction) would in practice be long (> 10 years). Phytoextraction cannot easily be combined with Allotment Gardens and is only a viable option if it could be safely designed and implemented to avoid potentially increased risks to human health, or grazing wildlife, due to possible contaminant uptake in edible crops grown on site (indicated by the red dotted arrow in Figure 4-3). An Allotment Garden land use with restrictions regarding crop selection and implementing safe agriculture practices could be a more feasible option in combination with using soil amendments with low- or non-accumulating plants to stabilise/immobilise the contaminants in the soil matrix and reduce bioavailability to prevent uptake into plants. However, it would require control of user's behaviour at the site, which in practice may be difficult.

4.4.2 Kalleberga

Given its current and expected future land use, the risk management framework has been applied at Kalleberga for only one green land use, a tree nursery, which is essentially equivalent to a Biofuel Park as modelled in the SEPA guideline value model (Figure 4-4). Instead of the SGVs created using generic assumptions, the site-specific guideline values generated by Tyréns for DDT were incorporated into the framework to better account for site-specific risk conditions. The dominating human health exposure pathway(s), or most sensitive environmental receptor(s), per contaminant and land use are indicated and linked with the corresponding risk mitigation mechanisms and potential GRO strategies. The calculated risk quotients are shown in the figure, and a $RQ > 1$ indicates an elevated risk (i.e. above the site-specific SGV). Accordingly, DDT contamination in concentrations measured at the site can be deemed to pose a potential risk primarily to the environment ($RQ = 7.25$, primary receptor: soil ecosystem) and not human health ($RQ = 0.45$), which aligns with the risk assessment performed by Tyréns (Sandström et al., 2020). The GRO strategies that are identified to be able to mitigate the dominating exposure pathways are highlighted in green boxes in Figure 4-4.

The risks posed to the soil ecosystem are of primary concern and GRO strategies can be applied to mitigate this risk by 1) reducing the bioavailability and consequent exposure for soil organisms by using e.g. soil amendments, and 2) removing the source of the contamination by either phytoextraction or phyto-/rhizodegradation. A combination of these strategies could reduce the exposure risks in the short-term (stabilisation/immobilisation) and/or achieve source removal in the longer term (extraction, degradation). The risk of DDT spreading to the groundwater could also be managed through the use of vegetation cover to limit infiltration of water through the soil profile thereby reducing leakage and providing hydraulic control. Thus,

a combination of different GRO strategies, a multi-mechanism application, could be the optimal solution for Kolleberga. In its current state, the agricultural fields remain largely unused with no immediate plans for redevelopment other than potential re-use as a tree nursery at some point in the future. Therefore, gradual removal of the source term via extraction or degradation could be well-suited to this site since there is no time constraint and the risks are relatively low and feasibly managed using GRO. Cultivating crops with potential economic benefits could further improve the value proposition of phytomanagement at Kolleberga.

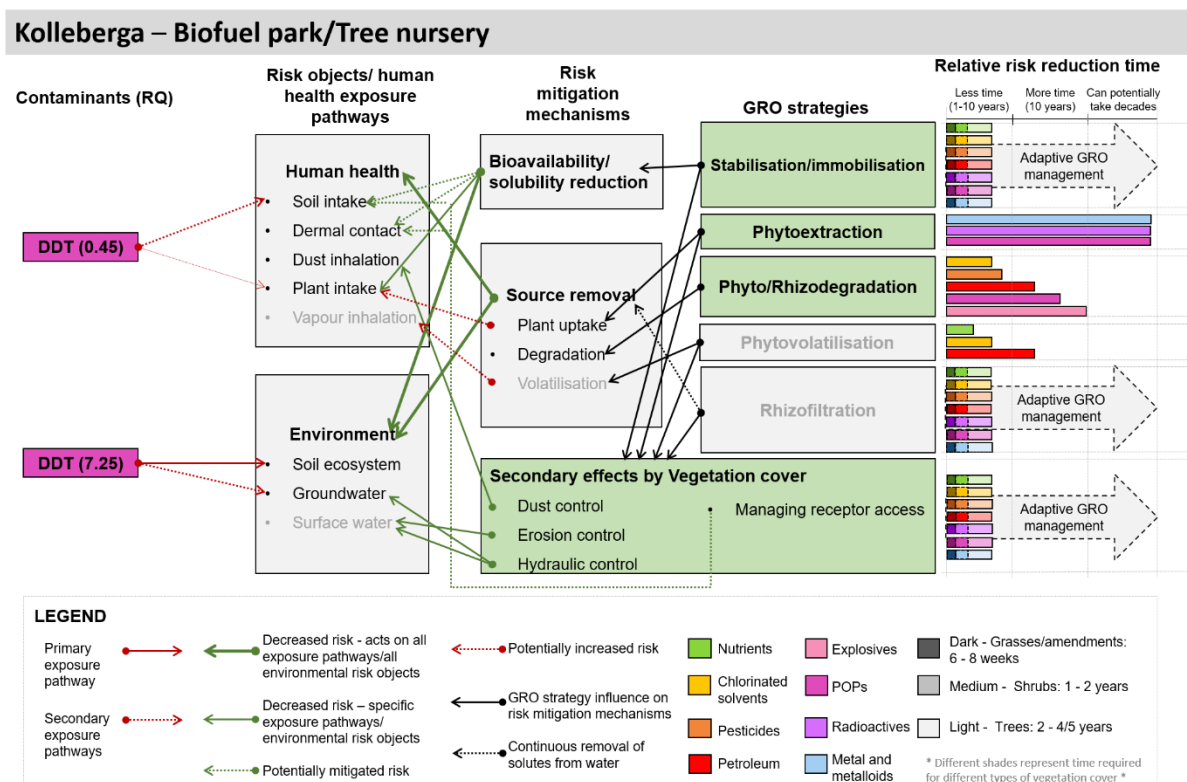


Figure 4-4. Site-specific application of the GRO risk management framework for the green land use of Biofuel park or Tree nursery. The contaminants detected at the site, Kolleberga, and risk quotients (RQ) are included in the furthest left column and are separated into exposure pathways for human health (above) or for the environment (below).

5 DISCUSSION

This chapter of the thesis presents a summary discussion on the thesis output and limitations as well as some broader implications concerning sustainable remediation and development, GRO, and phytomanagement application in Sweden.

5.1 GRO effectiveness, time perspective and application

The effectiveness of GRO as a remediation technique, for which applications it is truly feasible and how successful application is defined are key aspects determining its adoption in practice. It is difficult though to generalise regarding both remediation effectiveness and the expected timeframe as it is highly variable and dependent on site-specific factors. For organic contaminants, GRO degradation mechanisms have been shown to be highly effective for many contaminants and could reduce risks directly by source removal over a shorter time. However, effectiveness of phyto- and rhizodegradation is variable and depends on factors such as the type of organic compounds present, bioavailable and total concentrations, soil type, weathering of contaminants and plant species and tolerance amongst others (Mench et al., 2010; OVAM, 2019). Reports of organic contaminant removal rates by established, mature phytoremediation systems are rare in the published scientific literature; however, there have been a few studies that estimate removal rates of various petroleum products and chlorinated solvents by trees through a combination of extraction, degradation and volatilisation (e.g. (Andrew James et al., 2009; Cundy et al., 2020; Doucette et al., 2013; Gobelius et al., 2017; Lewis et al., 2015; Limmer et al., 2018)). In the case of highly volatile organic contaminants (VOCs), eventual volatilisation by plant transpiration that releases VOCs into the atmosphere could potentially increase the exposure risks in some situations (OVAM, 2019), which is a non-trivial possibility that must be accounted for in GRO design and monitored accordingly.

Regarding inorganic contaminants, the time expectations for phytoextraction are typically compared to that of conventional remediation options to be considered commercially viable; meaning that it should be completed within a 'reasonable timeframe' (e.g. <10 or <25 years) (Gerhardt et al., 2017; Robinson et al., 2006, 2003b; Van Nevel et al., 2007; Vangronsveld et al., 2009). Estimating the time required for phytoextraction, which can potentially take up to a few decades, is thus a critical aspect of determining the feasibility of phytoextraction. Due to inherent inefficiencies that typically result in a long remediation time-frame, phytoextraction with the narrow focus of exclusively taking up metals as a stand-alone technology may indeed rarely be suitable for strictly remediation purposes (Dickinson et al., 2009; Robinson et al., 2015, 2006; Van Nevel et al., 2007). However, alternative phytoextraction strategies like soil polishing (reducing marginally elevated concentrations to threshold levels) and bioavailable contaminant stripping (reducing the soluble, plant-available fraction of metals) are viable niche-solutions which could be more widely applicable at various scales and shorten remediation times from decades to just a few years (Dickinson et al., 2009; Gerhardt et al., 2017; Herzig et al., 2014; Mench et al., 2010; Robinson et al., 2015, 2009, 2006; Van Nevel et al., 2007; Vangronsveld et al., 2009). GRO strategies not purposed for source removal such as phytostabilisation or in-situ immobilisation could also be used to significantly reduce the bioavailability and solubility of (in)organic contaminants in a relatively short time. The vegetation cover itself controls erosion, dust and groundwater hydraulics to physically reduce

the risks and manage the receptors. Revegetating a contaminated site shows great potential for establishing either a long-term succession of plant communities or a sustainable cropping rotation that promotes soil development processes, enhances nutrient cycles, stabilises microbial communities and maintains sustainable soil ecosystem functions with either no or acceptable residual contaminant linkages (Cundy et al., 2016; GREENLAND, 2014a; Mench et al., 2010).

As shown in Table 4-3, GRO can be applied to manage risks in a variety of situations. It may also be possible to enable concurrent or future, more sensitive land uses by applying GRO over time and re-evaluating the risks. Depending on the site conditions and land use, exposure risks like possible human exposure due to plant intake necessitates caution and more in-depth risk assessment before sensitive land uses are validated on contaminated sites. When food crops are considered for cultivation on a contaminated site as in, for example, the studied allotment garden scenario. It is, however, possible to safely cultivate food crops in contaminated soils by either i) selectively cultivating crop varieties or clones that exclude (i.e. do not take up) contaminants from their edible biomass, ii) pre-cultivating or co-cropping contaminant accumulating (i.e. extractive) species with non-accumulating or excluding food crop varieties to further reduce plant uptake in food crops, or iii) pre-cultivating contaminant accumulating species to strip the bioavailable fraction and reduce contaminant uptake in subsequent crops (GREENLAND, 2014b; Greger and Landberg, 2015; Haller and Jonsson, 2020; Kidd et al., 2015; Tang et al., 2012). If growing food crops in the contaminated soil is still considered to pose an unacceptable exposure risk, then vertical systems could be used alongside safe agricultural practices and institutional controls (US EPA, 2011). Contaminant uptake into plants (or mesofauna) could also potentially increase the risk of exposure for grazing or predatory wild animals, but this risk can be effectively reduced through careful GRO site design and other engineered solutions to reduce access to contaminated areas in collaboration with stakeholders. GRO could be employed to enable simultaneous land use by tailoring the vegetation to stabilise contaminants in the soil matrix thereby preventing spreading by leaching, dust and erosion. Soil amendments could be used to enhance this effect while also providing a barrier between humans and the soil to further limit exposure risk by ingestion, dermal contact or dust inhalation. This strategy could be especially beneficial in urban gardens, for example, where it has been shown that the primary exposure pathways for humans to As and Pb are soil and dust ingestion, rather than vegetable consumption (Paltseva et al., 2020).

5.2 Possibilities and challenges for applying GRO in Sweden

Many of the reflections by the interviewed experts reaffirmed those acknowledged in other studies that address the various obstacles and limitations of GRO, perceived and actual. Broadly, these include a 'status quo bias' and preference for conventional methods like dig-and-dump by practitioners (Montpetit and Lachapelle, 2017); 'nonknowledge' by practitioners regarding their functionality, methods and dealing with uncertainties, limitations or inefficiencies in GRO application (Bleicher, 2016); ecological risks from secondary poisoning due to wildlife grazing on metal-enriched plants or the improper handling of harvested biomass that may have higher concentrations of risk elements (Dickinson et al., 2009; Wang et al., 2019); and other practical challenges and limitations such as uncertainties relating to the

required timeframes for GRO and their effectiveness as risk management strategies, applicability for different types of sites and contaminants, insufficient knowledge and experience, need for long-term monitoring, and lack of convincing proof-of-concept amongst other concerns (Cundy et al., 2016; Gerhardt et al., 2017).

In the Swedish context, lack of awareness surrounding GRO, relevant techniques and their effectiveness has been identified as a major obstacle to their implementation. A recent survey study, carried out as part of a bachelor's thesis project, investigated the awareness of GRO across the Swedish public and private sector (called nature-based methods in the study) which had 153 respondents in total (Berghel et al., 2021). The study revealed variable knowledge of three GRO with the majority of respondents (57%) aware of phytoremediation but fewer indicating awareness of the use of soil improving amendments (44%) or mycoremediation (27%) as remediation alternatives. Adoption of GRO is low in Sweden, with respondents able to identify only 8 projects (67% knew of none) in total where they were applied, and there was a shared uncertainty as to whether there would be an increased usage in the future. Most respondents viewed the time required, uncertainty and lack of knowledge as well as lack of consultants and contractors offering GRO commercially as the main obstacles to their implementation in Sweden, which aligns with other studies reporting GRO challenges. Likewise, the main advantages of GRO cited by respondents are that they are environmentally friendly, resource efficient and can support the recovery of ecosystem services (Berghel et al., 2021).

The inherent multi-functionality of GRO and its co-benefits are highly touted and recognized by experts and other remediation practitioners, which should improve its value proposition to stakeholders and decision-makers. All the interviewed experts, and many survey respondents, echoed the possibilities of wider benefits which begs the question 'why are GRO not used more often? Often, these added benefits are not accounted for in the decision-making process. Also, conservative regulatory guidelines based on total concentrations and full source removal could make the risk reduction via GRO prohibitively difficult to demonstrate. Therefore, a shift in perspective of managing contaminated sites is required. Not least in accounting for bioavailability in site assessment as a standard that is accepted by regulatory agencies, but also reformulating the remediation objectives in terms of 'upgrading degraded land' and 'risk reduction and management' instead of 'full source removal and decontamination'. Effectively valuing the benefits of GRO, accounting for them during options appraisal and raising GRO as viable remediation techniques are key aspects to their broader integration as viable land management strategies.

5.2.1 Implications for phytomanagement

As previously discussed in Drenning et al. (2020) (Paper I), transitioning from predominantly grey, 'hard' built infrastructure to 'soft' nature-based solutions (NBS) (Keesstra et al., 2018b; Song et al., 2019) or green infrastructure (Olofsdotter et al., 2013; Sandström, 2002), which both emphasise the multi-functionality offered by green spaces and natural processes, is considered to be essential to achieve the SDGs. Phytomanagement can play a key role in this transition. The economic benefits of phytomanagement are undoubtedly important for long-

term sustainability; however, the wider environmental benefits generated in phytomanagement, especially at larger sites, are becoming increasingly salient in the modern context of widespread environmental degradation, biodiversity loss, rising sea levels, climate change and other challenges to meet the Sustainable Development Goals (Bardos et al., 2020a; Keesstra et al., 2018a, 2016; O'Connor et al., 2019). Also, when viewed in this broader context as a nature-based solution (NBS), phytomanagement may gain wider acceptance as a mainstream land management strategy for broader situational applicability to contribute to sustainable development (Bardos et al., 2020a; Cundy et al., 2016; Keesstra et al., 2018b; O'Connor et al., 2019; Song et al., 2019). Especially now, as we enter the UN Decade on Ecosystem Restoration, phytomanagement can play a valuable role in 'upgrading degraded land' and achieving the EU goal of 'degradation neutrality' to preserve and restore land and soil resources that provide critical ecosystem services.

Long-term monitoring is another key aspect to evaluate the effectiveness of GRO for both ensuring regulators that contaminants are being managed as well as improving soil quality by monitoring important soil parameters linked to key soil functions or ecosystem services (Birgé et al., 2016; Burges et al., 2018; Epelde et al., 2014a; Garbisu et al., 2011; Gómez-Sagasti et al., 2012). Adaptive maintenance and monitoring (i.e. programs evolving iteratively to continuously improve) can be incorporated into phytomanagement projects in order to reduce uncertainty regarding remediation effectiveness and responses by soil biota to management (Birgé et al., 2016; Chapman, 2012; Epelde et al., 2014a). By including iterative decision points (e.g. every 5 years), it is also possible to re-examine the risk assessment at the site after a period of phytomanagement to determine whether the site is fit for a different type of land use (potentially more sensitive) that was previously excluded given the prior risk assessment. As noted in Chowdhury et al. (2020), alternative green land uses with various degrees of permanency are made possible over time with GRO interventions.

5.2.2 Integrating ecosystem services

There seems to be a general consensus that when a land (or soil quality) management strategy incorporates the concept of ecosystem services (ES), quantifiable soil features can be more easily linked to land-use expectations and protection targets in a defensible and transparent way (Bünemann et al., 2018; Burges et al., 2018, 2016; Epelde et al., 2014a, 2014b; Faber et al., 2013; Faber and Van Wensem, 2012; Garbisu et al., 2011; Gómez-Sagasti et al., 2012; Gutiérrez et al., 2015; Pulleman et al., 2012; Rutgers et al., 2012). The ES concept is becoming mainstream in policy and planning for communicating about the environment and operationalisation has even led to gradual changes in decision-making and action (Dick et al., 2018); however, in the context of contaminated soils, application of ecosystem service assessment or valuation methods is still limited to only a few studies (De Valck et al., 2019). ES can be integrated into soil/site assessment in various ways based on qualitative, semi-quantitative and/or quantitative methods, which then can be used to either assess the current state of a site or soil ecosystem, monitor the changing conditions over time or estimate/measure the change in ES resulting from a land management alternative (e.g. a remediation option) to support decision-making (Faber et al., 2013; Faber and Van Wensem, 2012; SEPA, 2018b; Volchko et al., 2020). Integrating ecosystem services and soil quality assessment using soil

quality indicators into contaminated site investigation, assessment and management is a significant step in the right direction towards sustainable soil and land management where soil is managed in accordance with the soil's capability and condition (Drenning et al., 2020; Volchko et al., 2019a). By accounting for soil parameters beyond just total contamination levels in decision-making (as is too frequently done in tier 1 risk assessments in Sweden), the latent potential of the soil can be leveraged to advance sustainable remediation and development (Volchko et al., 2019a). For, the ultimate objective of a risk-based and sustainable remediation process must be not only to remove the contaminants from the soils (or instead break contaminant linkages) but also to restore soil quality (Epelde et al., 2008; FAO et al., 2020; Gómez-Sagasti et al., 2012).

5.3 The risk management framework as a tool for communication

Many studies have reported the lack of knowledge amongst stakeholders of GRO generally and of currently available decision-support tools (DST) for brownfield redevelopment and GRO application, so a clear goal is to raise awareness and inform stakeholders of available DST and the viability of GRO for risk management (Berghel et al., 2021; Bert et al., 2017; Cundy et al., 2016, 2015; Gerhardt et al., 2017; GREENLAND, 2014b; Onwubuya et al., 2009). There are several DST already developed for GRO, which are often focused on the technical application details and practical considerations for designing the remediation strategy for a site, e.g. (Andersson-Sköld et al., 2014; Cundy et al., 2015; ITRC, 2009; Onwubuya et al., 2009; OVAM, 2019). These DST can be viewed as complex by decision-makers and may not be well-suited as a communication tool in the early stages of a site remediation project since they require knowledge and in-data that stakeholders may not have available. The risk management framework can thus function as a complement to (not a replacement for) existing DST for GRO implementation as it is targeted towards educating stakeholders – who are not necessarily trained in GRO and risk assessment – on the connections between risk mitigation mechanisms, risk objects, and GRO strategies. The risk management framework for GRO is intended to support remediation contractors, decision-makers, regulatory bodies and other stakeholders involved in contaminated sites. It can be used in the early stages of a brownfield redevelopment project as a tool for 1) communication of risk mitigation mechanisms by GRO and their associated timeframes, and 2) identifying opportunities for GRO implementation at specific sites preceding necessary site-specific risk assessments. In so doing, it can be a useful tool to address some of the concerns and fill some of the knowledge gaps identified by the expert group during interviews. For example, the viability of GRO as effective risk management strategies was questioned by interviewed experts and survey respondents. Using a pedagogic framework for communication can aid in alleviating these concerns by educating and informing stakeholders about the range of possible applications of GRO for effective risk management that can be customised along contaminant linkages to act on source, pathway and/or receptor.

Risk communication is a fraught topic that would benefit from a clear, transparent framework, in line with existing regulations, to use in the early stages of planning for brownfield redevelopment for discussing the wide variety of contaminant linkages that can be managed using GRO (Cundy et al., 2015; GREENLAND, 2014b; Hammond et al., 2021; Onwubuya et

al., 2009). As demonstrated in the case study applications, the risk assessment changes significantly depending on the desired end use and the framework strengthens the decision basis by clarifying relevant risk mitigation mechanisms to manage contaminant linkages and corresponding GRO strategies. A recurring debate in GRO application is regulator acceptance regarding managing risks without necessarily reducing total concentrations (i.e. source removal) and time concerns (Cundy et al., 2016; Gerhardt et al., 2017). The relative time for risk reduction in the generic framework is a simplified generalisation but it provides transparency with respect to the effectiveness of GRO strategies relative to time. This in turn can provide a starting point for setting reasonable expectations and communicating with stakeholders.

5.4 Limitations and possible developments of the framework

Perhaps the main limitation in the risk management framework is the rough generalisation of 'relative risk reduction time' that is inherent to GRO. Estimates for certain source removal mechanisms could be more easily gained from the literature (Kennen and Kirkwood, 2015; OVAM, 2019), however, risk reduction measures that focus on more complex soil chemistry dynamics like bioavailability reduction are more difficult to estimate and vary with site-specific conditions. For this mechanism, as well as vegetation cover, the time estimate was instead based on the approximate time for vegetation establishment or amendment activation to alter the soil environment. Future research to provide models enabling better prediction of the time required for the various GRO mechanisms would allow for greater sophistication in designing phytomanagement strategies to achieve an envisioned land use within a certain timeframe, though this would also require extensive monitoring and long-term field trials.

Furthermore, the various risk mitigation mechanisms and how they reduce exposure to risk objects were based on available literature and created to generalise the GRO strategies included in this framework. From the literature review, no supporting evidence could be found on whether a lower bioavailability would reduce the human uptake of contaminants via dermal contact, but it is typically not a dominating exposure pathway for most contaminants. Also, there is no consensus for measuring and including bioavailability in existing risk management frameworks (Kumpiene et al., 2017). In addition, the evidence for the reduction of contaminant oral bioaccessibility in the gastro-intestinal system via GRO is controversial, with conflicting results, and would require further examination to be considered a viable strategy (Gray et al., 2006; Mench et al., 2006; Paltseva et al., 2020; Pelfrène et al., 2015; Sanderson et al., 2015).

The background parameters and generic assumptions built into the SEPA model used to derive the SGVs in the framework are inherently conservative. Conservative assumptions in such risk models are common and most models account for similar exposure pathways. However, such conservatism does not easily allow for integration of bioavailable contaminant concentrations that is typically a basis for site-specific risk assessments. There is a need for standardised methods and clear knowledge for how it can be done so that it is recognised by regulatory agencies, which is particularly crucial in Sweden. Also, more in-depth knowledge would facilitate refining the parameters and assumptions used in the SEPA model to create more realistic exposure scenarios for the green land uses.

There is of course room for improvement in the framework and the most obvious would be to include bio-, myco-, vermiremediation as GRO strategies. These were omitted in the initial version of the risk management framework in favour of phytoremediation techniques, but they could fit into future versions as source removal strategies. Also, additional risk objects such as grazing wildlife would be useful to improve the risk assessment. Grazing wildlife share many of the same exposure pathways as humans at contaminated site, so in effect GRO could be used to mitigate risks to both receptors though this should be better shown in the framework.

6 CONCLUSIONS

The final chapter of the thesis presents a summary of conclusions from the thesis.

The main conclusions from the licentiate thesis are summarised below.

- Brownfields present a significant opportunity for advancing sustainable remediation and development in and around cities to meet national and international environmental goals, including at least UN Sustainable Development Goals 11, 13 and 15. In suitable situations, well-designed GRO strategies have the potential to play a significant role in long- and short-term risk-based land management at contaminated sites. GRO can provide many desirable co-benefits, restore soil functioning and enhance ecosystem services as elements of green infrastructure and nature-based solutions.
- GRO are highly applicable for large land areas and peri-urban areas which tend to not be profitable for 'hard' redevelopment and lie abandoned as conventional remediation techniques are unreasonably expensive or undesirable. There are many possibilities for applying GRO in Sweden and elsewhere with a perceived trend towards increased usage in the future. Many challenges remain to be addressed, including lack of awareness and knowledge of GRO by experts, contractors and decision-makers, the potentially long time required, shifting regulatory emphasis from total contaminant concentrations to bioavailable contaminant fractions and other general uncertainties regarding implementation, time requirements, effectiveness and successful remediation via GRO.
- Improved communication is needed to support risk reducing strategies that emphasise a risk-based perspective instead of focusing exclusively on total amounts of soil contaminants. The risk management framework is expected to facilitate better understanding and communication of the risk mitigating mechanisms and required timeframes of various GRO to support remediation contractors, decision-makers, regulatory bodies and other stakeholders related to contaminated sites. The preliminary risk reduction timeframes built into the risk management framework are derived from literature, very generic and broadly identified for groups of contaminants but could still be of considerable use to decision-makers.
- The case study applications demonstrated that an envisioned land use, site-specific contaminants and indications of the important contaminant linkages can be integrated into the generic framework to support the identification of relevant GRO strategies and also provide preliminary timeframes for risk reduction. The framework can thus act as an early-stage decision-support tool to educate and engage remediation contractors, decision-makers, regulatory bodies and other stakeholders related to contaminated sites to identify relevant GRO, including potential phytomanagement strategies. By including iterative decision points (e.g. every 5 years), it would be possible to re-examine the risk assessment at a site after a period of phytomanagement to determine whether the site is fit for a different type of land use that was previously excluded given the prior risk assessment.

7 ONGOING AND FUTURE WORK

This chapter identifies issues that require further investigation for GRO implementation and improving ecosystem services at contaminated sites as well as reflections, further steps and the ongoing work addressing these issues.

Ongoing and future work in this Ph.D.-project aim to address the following needs and knowledge gaps aimed primarily towards practical application of current research and best practices:

1. Field experiment: the three GRO strategies identified as suitable for DDT remediation at Kalleberga in Figure 4-4 are implemented in a pilot study to increase knowledge about GRO applications in the field and generate data for input to other studies. The strategies are 1) aided phytostabilisation using a) a grass mix and b) willow trees, 2) phytoextraction of DDT using pumpkin (*Cucurbita pepo*), and 3) rhizodegradation using the leguminous species clover and alfalfa/lucerne. All four treatments are tested with and without the addition of biochar.
2. Site investigations and assessment: building on the SF Box method (Volchko et al., 2019b, 2014b) to better integrate soil quality indicators and soil function/ecosystem service assessment into contaminated site management. Biological indicators that meet criteria for use in Sweden (e.g. cost, ease of sampling/understanding, commercial availability) will be tested and evaluated to provide guidance for practical application.
3. Time estimates: better estimates of the required timeframe for GRO have been identified as a key need, and preliminary work has been done to develop a tool to estimate the time required for phytoextraction. The preliminary model will be refined and data gained from the field experiment will be used as input for demonstration purposes.
4. Options appraisal: cost-benefit analysis (CBA) to compare GRO to conventional remediation techniques as basis for decision-making and options appraisal. Application of CBA for options appraisal aim to better integrate ecosystem services and other added benefits in the decision basis and the pilot study is planned to be used for demonstration purposes.
5. Guidance for working with GRO in Sweden: compiling knowledge gained and practical considerations for working with GRO in Sweden to create guidance for a practical working processes directed towards practitioners. The risk management framework will be integrated as a key part of the working process.

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9 PUBLICATIONS